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## Variation in chemical composition of MSWI fly ash and dry scrubber residues

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## ABSTRACT

Our society generates extensive amounts of municipal solid waste (MSW), which are mainly incinerated for volume reduction and energy recovery. Though, MSW incineration generates hazardous air pollution control (APC) residues that must be treated and deposited in appropriate landfills. An alternative to landfilling is material recovery, leading to regeneration of valuable products and reducing hazardous waste amounts. The chemical composition of APC residues, stemming from MSW, makes the waste attractive for metal and salt recovery, but its variation makes the development of material recovery processes challenging. This study investigates results from 895 X-ray fluorescence analyses of fly ash and dry scrubber residue samples originating from Norway and Sweden between 2006 and 2020 to explore variation in chemical composition within and between different incineration plants. The average relative standard deviation of elemental concentration in APC residue was estimated to 30% within plants. The variation in elemental concentration between grate fired incineration plants is about half of the average variation within the plants. The study also clarifies compositional differences from APC residues originating from fluidized bed incinerators and grate incinerators. Also, reported concentrations of APC residues from other countries than Sweden and Norway showed significant differences in chemical composition. The presented variations clarifies the importance of holistic approaches for waste valorization processes which can substitute stabilization processes for landfilling.

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### 1. Introduction

With the increase in municipal solid waste (MSW) generation and the lack of raw material availability in Europe, the importance of MSW management and material recovery is imperative. According to Eurostat, Norway and Sweden generated 739 kg and 434 kg MSW per capita in 2018, respectively. This makes Norway among the largest producers of MSW in Europe, whereas Sweden lies at the European average. MSW incineration (MSWI) has become the

standard method for treating MSW with its benefit of 95% volume reduction and additional energy recovery. During the incineration process, volatile metals, halogenide compounds and persistent organic tend to mobilize into the flue gas (varying with their volatile character). Consequently, solid matter originating from the flue gas cleaning system is high in concentration of toxic pollutants and is therefore deposited in appropriate landfills.

The International Ash Working Group (IAWG) (1997) defined fly ash (FA) as collected flue gas dust prior to any sorbents and reagent injection in the flue gas cleaning system. In comparison, air pollution control (APC) residues are defined as residues originating from dry scrubbers, semi-dry scrubbers and wet scrubber systems. In this article, the definition presented by IAWG is used for designating FA, dry/semi-dry scrubber residues are referred to as DS residues, and APC residues is used as a collective term for all solid residues from air pollution cleaning systems. The term APC residues is also used to design residues that are a combination of FA and DS residues or used when there are uncertainties about the residues' origin. FA usually originates from a wet-scrubber system

*Abbreviations:* APC, air pollution control residues; BFB, bubbling fluidized bed; CFB, circulating fluidized bed; DS, dry/semi-dry scrubber; EO, extreme outliers; FA, fly ash; FB, fluidized bed incinerators; FB-APC, air pollution control residues from fluidized bed incinerators; G, grate incinerators; G-DS, dry scrubber residues from grate incinerators; G-FA, fly ash from grate incinerators; IAWG, International Ash Working Group; MSW, municipal solid waste; MSWI, municipal solid waste incineration; RSD, relative standard deviation; SD, standard deviation; WS, wet scrubber; XRF, X-ray fluorescence analysis.

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where electrostatic precipitators remove all particular matter before the flue gas enters a wet scrubber. For grate incinerators, the acronyms G-FA and G-DS are used for FA and DS residues, respectively. For APC residues from fluidized bed incinerators, the acronym FB-APC is used.

Hazardous waste landfills may be potential sources of pollution. Because opening new landfills are costly and politically difficult, efforts have been made to valorize APC residues. Waste valorization and material recovery processes are good alternatives to land-filling with the benefits of reducing hazardous waste amounts and the generation of valuable products. Environmental benefits can justify the use of additional resources and costs. Quina et al., (2018) emphasize 4 different routes that promote recovery of secondary raw materials/products from APC residues: decontamination/detoxification, product manufacturing, practical application, and recovery of materials. All these routes are dependent on the characteristics of the APC residue, e.g. chemical composition, mineralogy, morphology and organic constituents. Huber et al., (2018) showed that a metal recovery process is the most environmentally friendly waste management strategy when comparing four different strategies in a life cycle analysis (LCA) study. One of the most uncertain parameters in the LCA model was the metal concentration in FA.

Hydrometallurgical methods have shown promising results for removal and recovery of toxic heavy metals. In Switzerland, Zn and Pb metal concentrate is recovered from FA with the FLUWA FLUREC process (Weibel et al., 2017). A large pilot plant is testing the HALOSEP® process operated by Stena Recycling A/S to recover salts at the Vestforbrændingen plant in Denmark (“Life HaloSep”, 2020). Both processes use spent acid scrubber liquor to leach metals from FA. The FLUWA FLUREC process uses metallic Zn cementation to remove less noble metal impurities and further liquid extraction and electrowinning for Zn recovery. HALOSEP® uses selective precipitation by regulating pH to separate heavy metals and a salt solution. Studies on Cu recovery utilizes solvent extraction and leaching with various solvents on Swedish fluidized bed (FB) incineration APC residues have also been performed (Karlfeldt Fedje et al., 2012; Tang, 2017; Tang and Steenari, 2015). Even though the methods mentioned above have demonstrated positive results for metal recovery, such processes can be sensitive to elemental compositional variations resulting in variable yields and challenging quality control. Elomaa et al., (2019) point out that Fe can be a costly impurity in their leaching study. The alkaline character of the residues can also offer challenges. The Ca concentration, as the largest constituent in APC residues, indicates the residues acid neutralization capacity, which is important for acid consumption when aiming for leaching at low pH. High acid consumption will be economically disadvantageous. However, leaching with concentrated solution may reduce costs by the lower total volume of aqueous solutions.

Removing highly soluble salts from APC residues might be the most uncomplicated process for material recovery, where residue reduction and reduced heavy metal mobility in deposited residues are gained. Here, Colangelo et al., (2012) have studied water consumption for washing out salts from APC residues. Further, utilizing APC residues for construction materials, which demands large amounts, would significantly decrease the volume needed for applicable landfills. Ca, Si, Al, and Fe phases present in APC residues make the residues applicable as raw materials in the cement industry (Quina et al., 2018). Here, chlorides are undesirable, but sulphates are acceptable (Moen, 2019).

The dynamic character and variation in APC residues' chemical composition make the development of recycling and material recovery processes challenging. Such processes must be robust and withstand variations in chemical composition. The elemental composition varies with geographical regions and time (year and

seasons) (Haberl et al., 2018; Ohbuchi et al., 2019; Wang et al., 2019). It also varies with incineration technology (Leckner and Lind, 2020), flue gas cleaning system (Jurczyk et al., 2016a, 2016b), upstream waste management system and downstream management (e.g. mixing of different APC fractions for storing and transport). Metals have different partitioning between bottom ash and APC residues. The partitioning is dependent, e.g. on strength of convective air flows, on vaporization of metals which again depends on temperature, the residence time in the incinerator and MSW composition that can catalyze vaporization reactions. Ruth, (1998) reports the enrichment factors (concentration in FA compared to the concentration in waste fuel) for selected metals: Cd 500, Sb 60, Zn 25, Pb 20 and Cu 3 for grate fired incinerators. The enrichment factors are significantly different in FB incinerators because the chemical conditions for gaseous species formation are different. Also, the total mass partitioning between APC residues and bottom ash is different. About 3–4% of incinerated waste ends up as APC residues and 15–20% as bottom ash in grate incinerators. In comparison, FB incinerators lead to about 8–9% APC residues and 6–7% bottom ash (Olofsson, 2006). According to Olofsson (2006), more APC residues are generated from FB incinerators, but the solid residues' total mass is lower. Different scrubber designs with variations of reagents and adsorbents generate concentration variation. Activated carbon is usually added to remove organic pollutants and Hg prior to dust filters (Jurczyk et al., 2016b).

This article provides an overview of 895 X-ray fluorescence analyses (XRF) samples of DS residue and FA from plants in Norway and Sweden gathered from 2006 to 2020. Variation in chemical concentration of APC residues originating from incinerators with different combustion technologies and flue gas cleaning systems are explored and presented. Differences between FB-APC and G-APC, as well as differences between G-FA and G-DS, are quantified. The chemical concentrations are compared to residues from other countries for putting Scandinavian APC residue characteristics into a global perspective. We consider this important for clarifying variations that can affect holistic waste management and material recovery process strategies for APC residues. Understanding the variation of APC residues is key for developing robust material recovery processes that can handle variation in chemical composition.

## 2. Methodology

### 2.1. Data source

A data set of chemical analyses of MSWI APC residues performed by NOAH AS at the Langøya landfill in Norway is the basis for this study. NOAH AS has performed XRF of Swedish and Norwegian APC residues between 2006 and 2020. Samples are taken from incoming trucks to Langøya. These are considered representative of bulk APC residues and rather homogenous due to well mixing during the transfer from temporarily storage siloes to trucks for transportation. Most samples originate from yearly verification of toxic heavy metals and many from periods with intense XRF analyses for technology development. The number of samples per year does hence vary.

Table 1 gives an overview of the dataset. The data represent 127 000 tons per year of MSWI residues which is the amount of residues delivered annually from all plants considered in 2017–2020. The plants are denoted with the letters A-Z. There are 310 samples of G-DS residues (all lime-based), 165 samples of G-FA, 65 samples of G-APC residues and 355 of various FB-APC residues. Further, circulating fluidized bed (CFB) and bubbling fluidized bed (BFB) combustion technologies are differentiated, as for dry scrubber

**Table 1**

Overview of dataset, where FA is fly ash, DS is dry scrubber, SDS is semi dry scrubber, WS is wet scrubber filter cake.

Plant	MSWI incineration technology	Type of MSWI residue	# XRF samples	XRF sampling years	Yearly tonnage waste delivered to Langøya in the period between 2017 and 2020
A	Grate	DS	44	2006–2020	5800
B	Grate	DS	43	2005–2020	3600
C	Grate	DS	20	2007–2020	1200
D	Grate	DS	18	2006–2020	1600
E	Grate	DS	64	2006–2020	16,600
F	Grate	DS	18	2006–2020	1800
G	Grate	SDS	38	2015–2017	–
H	Grate	DS/SDS	18	2010–2020	2600
I	Grate	SDS	18	2011–2020	2700
J	Grate	DS/SDS	14	2009–2020	800
K	Grate	DS/SDS	15	2009–2020	3500
L	Grate	FA	30	2008–2020	2800
M	Grate	FA	41	2007–2020	13,700
N	Grate	FA	18	2006–2020	1400
O	Grate	FA	45	2007–2020	1900
P	Grate	FA	31	2006–2015	–
Q	Grate	SDS/FA	15	2006–2015	–
R	Grate	SDS/FA	38	2010–2020	6400
S	Grate	SDS/WS	12	2010–2020	1900
T	FB (CFB)	SDS	121	2014–2020	24,500
U	FB (CFB)	SDS	29	2015–2020	5800
V	FB (CFB)	SDS	40	2015–2020	7300
W	FB (BFB)	DS	28	2011–2020	6100
X	FB (BFB)	Unknown	72	2009–2020	8100
Y	FB (BFB)	DS	46	2007–2020	3300
Z	FB (BFB)	DS	19	2008–2020	4000

residues (DS) and semi-dry scrubber (SDS) residues. Elsewhere in this article, CFB and BFB are combined, and so are DS and SDS. Before 2017, samples were analyzed with a loose powder XRF while samples from after 2017 were analyzed with pelletized powder XRF.

## 2.2. Descriptive statistics

The residues from the different plants were grouped into DS from grate incinerators (Plants A–K, Table 1), FA from grate incinerators (Plants L–P, Table 1), mixed APC residues from grate incinerators (Plants Q–S, Table 1) and APC residues from FB incinerators (Plants T–Z, Table 1). Mean elemental concentration with standard deviations (SD) were calculated for each group after extreme outliers were removed. The extreme outlier criterion was:  $X < Q1 - IQR \times 3$  or  $X > Q3 + IQR \times 3$ , where X is the elemental concentration, Q1 is the lower quartile, Q3 is the upper quartile and  $IQR = Q3 - Q1$ . The normality of the distributions of the elemental concentrations for each group was checked visually with normal probability plots and tested with the Shapiro-Wilk test. A robust principal component analysis (PCA) was performed in order to see variations between the groups in a multivariate system. The software package R 4.03 with robustPCA from the package pcaMethods (v1.64.0) was used.

## 2.3. Variation due to MSWI technology

The mean elemental concentrations of MSWI APC residues originating from grate incinerators were compared with those originating from FB incinerators to explore differences emerging from different combustion technologies. The distribution in G-DS, G-DS, and FB-APC concentration are presented in density plots (kernel smoothed) in the results chapter. Wilcoxon tests were also executed, shown in the Appendix. The same procedure was followed for the mean elemental concentration of G-DS, which were compared with G-FA to compare APC residues originating from different flue gas cleaning systems.

## 2.4. Geographical variation

The XRF data from different MSWI technologies and flue gas cleaning systems were compared with literature data to highlight the geographical variations in chemical composition. Three different sources were selected for comparison: First a comprehensive overview of FA and DS residues from 1997 illustrating time variation (The International Ash Working Group (IAWG), 1997); secondly, the comprehensive overview of heavy metals from Chinese grate incinerator and FB incinerator APC residues (Wang et al., 2019); and third, studies related to the Zn recovery process FLUWA FLUREC in Switzerland (Weibel et al., 2018, 2017). The number of samples for each source is about 40 (varying with the element), about 200 (varying with the element) and four samples, respectively.

## 2.5. Other variations

Variations between plants that are not subjected to geographical or principal technological characteristics were explored. These variations originate, e.g. from the number of incineration lines at a plant, from changes in fuel composition in short and longer perspectives and from different designs of flue gas cleaning system. After removing extreme outliers (same as previous criteria) from samples originating from each plant within the groups (G-FA, G-DS, and FB-APC), the average standard deviation was calculated as a measure of the variation within plants. The average standard deviation in a group was compared to an estimate of the variation between the plants in the group. The latter estimate denoted  $\tau$  was obtained using a one-way random-effects ANOVA model, as explained in Appendix E. This was done in order to compare the variation of APC residues within and between plants.

Elements with a large potential for material recovery and elements with the most extensive variations in either technology, geography or time were presented for all statistical analyses. V, As, Ag, Co, Ni, and Mo had too low concentrations to be reliable for the XRF method and were therefore excluded from the analyses. The Appendix contains more comprehensive results, normal

probability plots, mean elemental concentrations for the plants, estimates of the variation between the grouped plants, and statistical hypothesis testing results.

### 3. Results and discussion

The mean concentration, standard deviation, relative standard deviation, number of removed outliers and the number of samples for selected elements are presented in Table 2. Fig. 1 B-D presents boxplots ordered from highest to lowest elemental concentration. Regarding the highest concentrations over 10 wt%, both Ca and Cl concentrations can be found in G-FA and G-DS (B and C), while only Ca can be found in FB-APC (D). Regarding concentrations between 0.1 and 1 wt%, Na, K, S, Si, Al, and Zn are found in G-DS and G-FA while Cl, Si, Al, Na, Fe, S and K are found in FB-APC. Elemental concentrations of Cu, Zn and Pb, are between 0.01 and 0.1 wt% for FB-APC. The Cu concentrations are lower than 0.01 wt% for G-DS and G-FA. Cu and Fe have the most extreme outliers for grate incineration residues, while Si, Al and S have the most outliers from FB-APC residues (Table 2).

The PCA (Fig. 1A) shows an apparent clustering of samples from the grouped incineration technologies and explains 67.3% of the variance (47.8% for the first principal component and 19.5% for the second component). The first principal component (PC1) separates the difference between G-APC residues and FB-APC residues, and a high score is decided by the high concentrations of non-volatile elements (Cu, Fe, Al, Ba, Si, Ti, Cr and P). FA and DS scrubber residues from grate incinerators are separated into two clusters by the second principal component (PC2). High PC2 scores are decided by high concentrations of volatile elements (e.g. Sb, Cd, S, Zn, K, Sn and Pb) and relatively lower Ca and Cl concentrations. In Fig. 1 A, green colour is G-APC residues not covered by G-DS and G-FA and represent plant Q-S in Table 1. These samples are more closely linked with DS residue than FA. The FB-APC residues clustering into several groups in the PC1 direction indicates a large variation in FB residues from plant to plant. This difference comes from the different FB combustion technologies (variations of CFB and BFB) and flue gas cleaning system.

Normal probability plots for the elemental distributions in the groups G-DS, G-FA and FB-APC and the results of Shapiro-Wilks test for normality are given in Appendix B. Most elemental distributions deviate significantly from the normal distribution. The most considerable deviations from normality are found in the FB-APC group, followed by the G-DS group. Elements with the most significant deviation from normality are Al, Ba, Fe, P, S, and Si for grate incinerators. The deviations are generally high for FB-APC elemental concentration. Density plots with mean concentration

marked are given for selected elements in Figs. 2 and 3 and for all elements in Appendix C. These figures show that mean concentration is a good measure of centrality for the distributions.

#### 3.1. Variation due to MSWI technology

##### 3.1.1. Combustion technology

FB-APC residues have significantly higher mean elemental concentrations of non-volatile metals compared to G-APC residues. Cu, Fe, Al, Ba, Si, Ti, Cr and P show higher concentration by the first principal component in Fig. 1 A. The mean elemental concentration ratios for FB-APC versus G-FA and G-DS are highest for elements in the following order: Cu (6.8 and 4.4), Fe (4.1 and 2.3), Al (4.4 and 1.8) and Ba (3.0 and 1.6). The differences are also apparent for Ti (2.9 and 1.2), Si (2.8 and 1.2) and Cr (2.1 and 0.9). In contrast, the elemental concentrations are much higher for volatile elements in G incinerators compared to FB incinerators. The mean elemental concentration ratios for G-FA and G-DS versus FB-APC are K (3.7 and 1.9), Cd (2.5 and 2.0), Na (3.2 and 1.9), Sb (2.5 and 2.0), Cl (1.7 and 2.1), Zn (2.3 and 1.6), Sn (2.0 and 1.3) and S (1.7 and 1.4). The differences are clearly seen in density plots in Fig. 2. Figures for all elements are found in attached Appendix C.

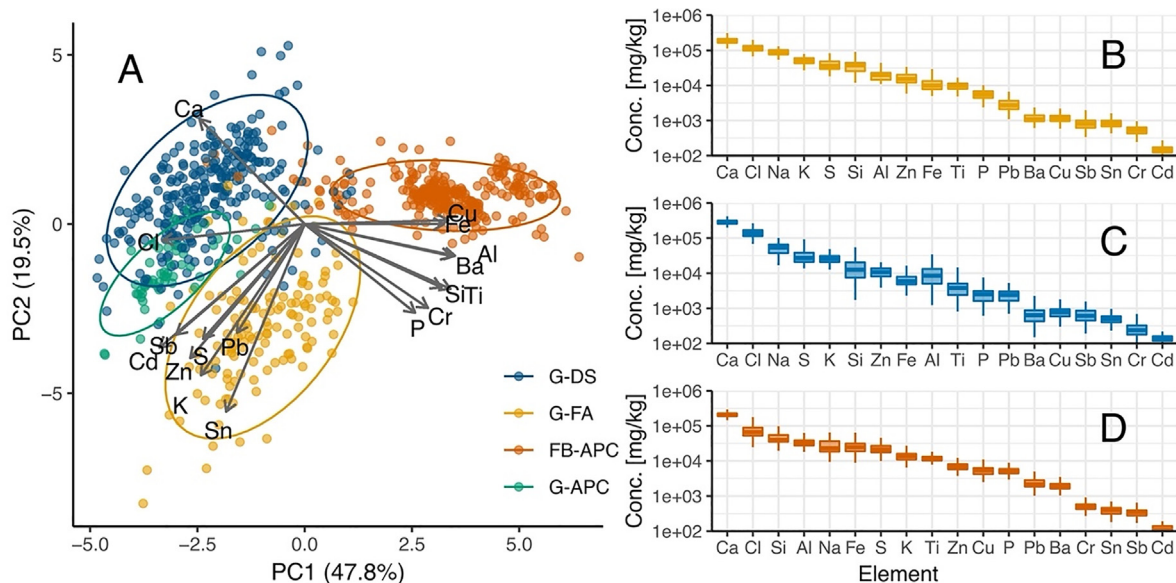
The difference between FB-APC and G-APC can be explained by the different elemental partitioning between bottom ash and APC residues. For FB combustion, a larger portion of residues ending up in the flue gas cleaning system is due to more turbulent conventions leading to higher concentrations of non-volatile elements in APC residues. Metallic Al concentrations of 4–5 wt% in Swedish CFB APC residues have been reported (Östrem, 2019). This indicates that a large portion of Al in FB-APC is in the metallic state. This can give rise to special residue properties as metallic Al has a high reduction potential. Some Si and Al may come from the bed material (sand) that end up in the flue gas.

Another key difference for the combustion technologies is the difference in chemical condition for volatilization. FB operates at lower temperatures than grate incinerators. Nonetheless, the operating temperature depends on the plant design and can vary significantly. One FB plant included in this study reports incineration temperatures around 500 °C. It is also reported that typical temperatures is 850 °C for FB incinerators compared to <1100 °C in grate incinerators (Leckner and Lind, 2020). Lower temperatures in FB incinerators result in lower up-concentration of volatiles. The vaporization kinetics and equilibrium are very dependent on temperature. Different kinetics and equilibrium below and above 750 °C have been reported (Falcoz et al., 2009; Rio et al., 2007). The low temperature in FB hinders heavy metals like Cd, Pb and Zn to be volatilized in contrast to grate incinerators which have

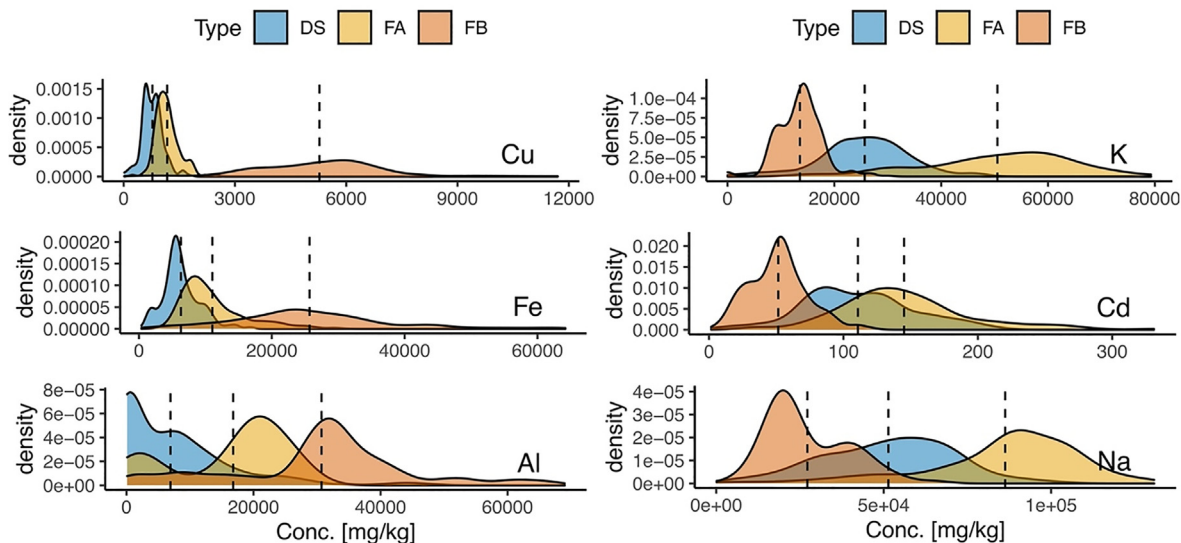
Table 2

Descriptive statistics of dataset. Concentrations are in mg/kg. SD is standard deviation, RSD is relative standard deviation, EO is number of extreme outliers and n is number of samples. Mean, SD and RSD is calculated from groups trimmed for EO.

		Cu	Pb	Zn	Cd	Sb	Sn	Cr	Fe	Ca	Si	Al	S	Cl	Na	K
G-FA	Mean	1200	2900	15,900	200	800	900	500	11,000	187,200	37,700	17,300	39,000	118,200	84,900	51,200
	SD	260	980	5250	50	270	200	130	3970	42,040	15,100	8730	11,790	27,100	19,260	12,530
	RSD	22%	34%	33%	25%	34%	22%	26%	36%	22%	40%	50%	30%	23%	23%	24%
	EO	8	1	0	2	2	0	0	4	1	0	0	1	1	0	0
	n	164	164	164	164	163	164	164	164	163	163	163	163	163	163	119
G-DS	Mean	770	2400	10,900	110	640	490	240	6300	288,500	15,500	6900	31,000	138,700	51,400	25,700
	SD	280	970	3900	40	280	160	130	2800	46,800	11,300	7400	13,400	43,900	19,200	8400
	RSD	36%	40%	36%	37%	44%	33%	53%	44%	16%	73%	107%	43%	32%	37%	33%
	EO	4	0	0	0	0	0	3	7	2	3	0	1	0	1	1
	n	305	305	305	305	305	305	305	305	305	305	305	305	305	305	164
FB-APC	Mean	5280	2270	6950	50	340	390	500	25,680	203,950	43,700	30,710	22,500	66,900	27,230	13,550
	SD	1670	930	1890	20	110	100	150	11,430	43,930	18,860	14,150	7590	36,370	12,310	4140
	RSD	32%	41%	27%	40%	32%	26%	30%	45%	22%	43%	46%	34%	54%	45%	31%
	EO	1	5	5	3	5	2	5	3	6	10	12	14	1	0	3
	n	355	355	355	355	355	355	355	355	355	355	355	355	355	355	164



**Fig. 1.** A: Principal component analysis of XRF dataset. B-D: Elemental concentration ordered from highest to lowest concentration for G-FA, G-DS and FB-APC, respectively. Note logarithmic y-axis. In figure A, green are G-APC residues which are neither pure DS nor FA, but rather a mix of different APC residues. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)



**Fig. 2.** Concentration distribution for elements with the highest difference between mean elemental concentration from G-APC and FB-APC, where Cu, Fe and Al are highest in FB-APC and K, Cd and Na are highest in G-APC. Stippled lines are concentration means.

higher combustion temperatures. From highest to lowest, Cd, Sb, Zn, Sn and Pb are the heavy metals with a significantly higher concentration in G-APC than FB-APC. This is the same order as elemental enrichment factor from waste to FA reported by Ruth (1998).

The alkali volatilization is very dependent on temperature (Ma et al., 2020) and alkali chlorides volatilize around 700 °C, whereas alkali carbonates and sulfates decompose around 850 °C. Char-bound alkalis also decompose significantly in the temperature range between 800 and 850 °C. The highest differences in elemental concentration between FB-APC and G-APC observed in this study are K, Cd and Na.

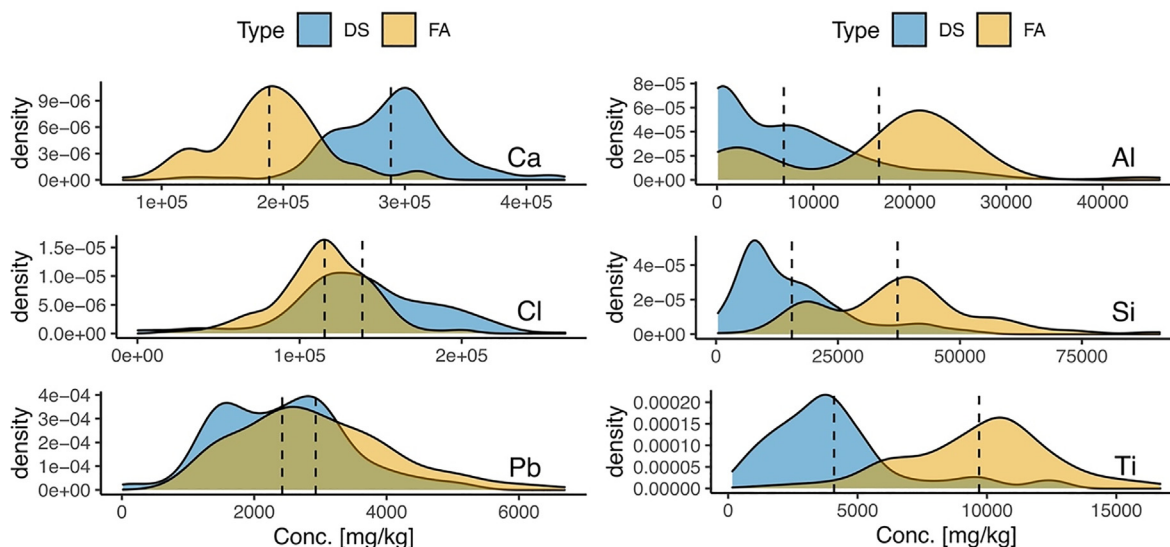
The high heating rate in FB incinerator retards Cl volatilization compared to the slow heating rate in grate incinerators (Liu et al., 2001; Ma et al., 2020). Experiments have also shown significantly less Cl and S volatilized at 600 °C compared to 900 °C (Watanabe et al., 2004). The difference in Cl concentration between FB-APC

and G-APC reflects that lower temperature in FB incinerators does not volatilize Cl to the same extent as grate incinerators. The Cl concentration is about twice as high for G-APC compared to FB-APC.

Because of Cr’s non-volatile character, a more considerable difference in concentration of FB-APC than G-APC is expected. This is, however, not observed in this study, which can indicate special Cr volatilization mechanisms in grate incinerators. Ebbinghaus (1995) show with thermodynamic modelling that Cr<sub>2</sub>O<sub>3</sub> can be volatilized as CrO<sub>2</sub>Cl<sub>2</sub>(g) with HCl(g) present at typical incineration conditions.

### 3.1.2. Flue gas cleaning system

Only the mean elemental concentrations of Ca and Cl are higher in G-DS than in G-FA: Ca is 1.5 times higher, whereas Cl is 1.2 times higher. The lowest mean concentration ratios of G-FA versus G-DS



**Fig. 3.** Concentration distributions for elements with highest difference between mean elemental concentration from FA and DS from grate incinerators. Stippled lines are mean concentrations.

are Ca (0.6), Cl (0.9), Pb (1.2), S (1.3), Sb (1.3), Cd (1.3) and Zn (1.5). The Cu, Mg, Na, Sn, Fe, Ba and K mean concentrations of G-DS versus G-FA lie between 1.5 and 2. The highest differences are present for P, Cr, Ti, Si and Al, which lie between 2.2 and 2.4 times higher. The elements with the most considerable difference between mean elemental concentration from FA and DS from grate incinerators are presented in Fig. 3. Figures for all elements are displayed in Appendix C.

The Ca mean concentration is approximately 100 g/kg higher in DS residues compared to FA. This indicates that 10% of DS residues is added lime. The difference in Cl content illustrates the DS system's efficiency for removing HCl(g) from the flue gas. The lime addition is primarily for HCl(g) neutralization. HCl(g) is stable at low temperatures, while other gaseous species (e.g. SO<sub>x</sub>, KCl(g), K<sub>2</sub>SO<sub>4</sub> (g)) nucleates before particle filters in wet scrubber systems (Ma et al., 2020). Therefore, the concentrations of S are not higher in DS residues as regular particle filters can remove most S-based gaseous species before wet scrubbers.

The difference in elemental concentration between the two different flue gas cleaning systems can indicate which elements have the highest chance of going through a particle filter without the addition of neutralization agent, adsorbents or other reagents. This may imply that Pb, S, Sb, Cd and Zn, in that order, tend to form stable gaseous species that can penetrate regular particle filters when no reagents or neutralization agent are added for nucleation.

### 3.2. Geographical variations

Boxplots presenting selected elemental concentrations from this study and literature data are shown in Fig. 4. Figures for all elements considered can be found in Appendix D while the mean elemental concentrations found in literature are presented in Table 3. The Zn and Pb concentrations are approximately twice and three times higher for G-FA reported by Weibel et al. (2018, 2017) than for G-FA analyzed in this study. In Switzerland, industrial waste is commonly mixed with MSW for incineration, which results in the elevated heavy metal concentration of the generated FA (Haberl et al., 2018). The high elemental Zn concentration makes the Swiss FA more suitable for Zn recovery with, e.g. the FLUWA FLUREC process. The Swiss FA is only based on 4 different samples, which question its representativeness.

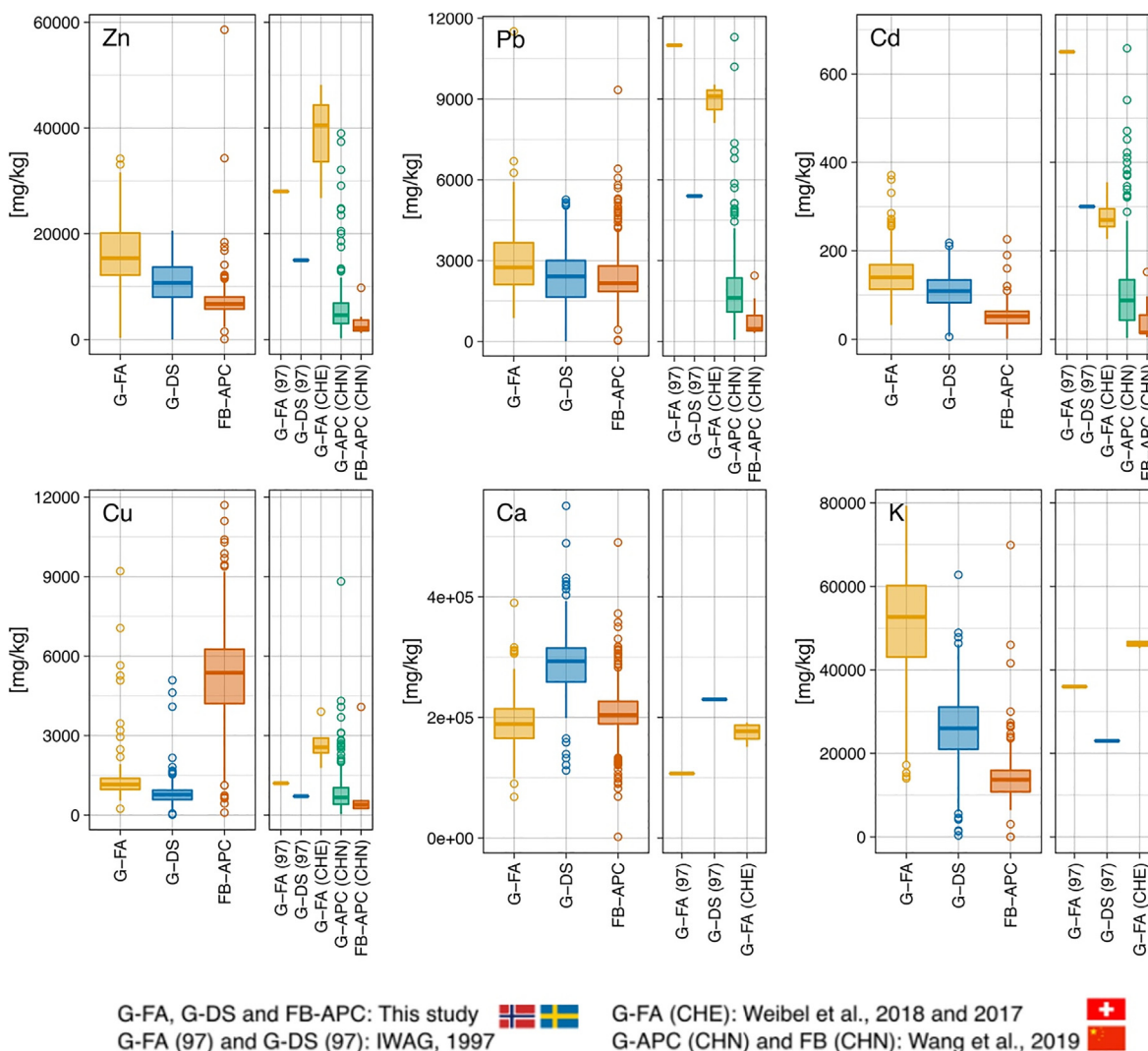
Chinese APC residues are significantly lower in heavy metals than Swedish and Norwegian APC residues. Even Cu and Cr concentrations from FB incinerators are lower than concentrations in G-FA and G-DS residues. The Chinese MSW composition differs from high-income countries where, e.g. the organic fraction is higher (Mian et al., 2017). The APC residue concentrations reported by IAWG (1997) are significantly different than the values reported in this study, in particular for heavy metals like Pb and Cd. This clarifies that the elemental concentration of APC residues changes with time due to waste fuel changes.

### 3.3. Other variations

The variation in chemical concentration from plant to plant and within plants can be seen in Fig. 5, where each plant's concentration distributions are presented in boxplots for selected elements. Figures for all elements can be found in Appendix E. The average relative standard deviation for the elemental concentration of G-DS plants is 28%; for G-FA plants, it is 33%, and for FB-APC plants, it is 22%. The average relative standard deviation for Ca in DS residues is about half of the other elements because Ca is added as the main element in lime-based neutralization agent in a controlled manner. The average RSD for all elemental concentrations for G-FA plants was 28%, close to RSD for all samples in the whole group (29%). The RSD for all samples in the G-DS group is 47% and is much higher than the average RSD within the plants at 33%. This indicates that there are more considerable differences between DS residues than FA residues from various plants.

The variation between plants estimated by  $\tau$  (Appendix B) showed equal trends. On average, the  $\tau$  estimates are 40% less than the average standard deviation for G-FA plants, while they for G-DS plants on average are 20% lower than the average standard deviation. This distinction might be explained by more considerable variations in dry/semi-dry scrubber designs compared to particular filter designs utilized before wet-scrubber systems. Also, the different reagent types may cause a larger variation for DS residues. The average  $\tau$  is 40% higher than the average standard deviation per incinerator for FB-APC plants, resulting from FB technologies (e.g., CFB and BFB) and flue gas cleaning system technologies not being differentiated.

The estimate  $\tau$  is obtained from a one-way random-effects ANOVA model. As seen in Appendix E, this model assumes equal



**Fig. 4.** Zn, Pb, Cd, Cu, Ca and K elemental concentration of APC residues from this study (left plot) and from the literature, Table 3 (right plot). G-FA, G-DS and FB-APC are represented by yellow, blue and red colours, respectively. In plots of literature data, green colour represents grate incineration APC residues. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

**Table 3**  
Mean elemental concentration in mg/kg of literature data where G-FA (97) and G-DS (97) are from IAWG (1997), G-FA (CHE) is Swiss FA from Weibel et al. (2018, 2017) and G-APC (CHN) and FB (CHN) are Chinese APC residues from Wang et al. (2019).

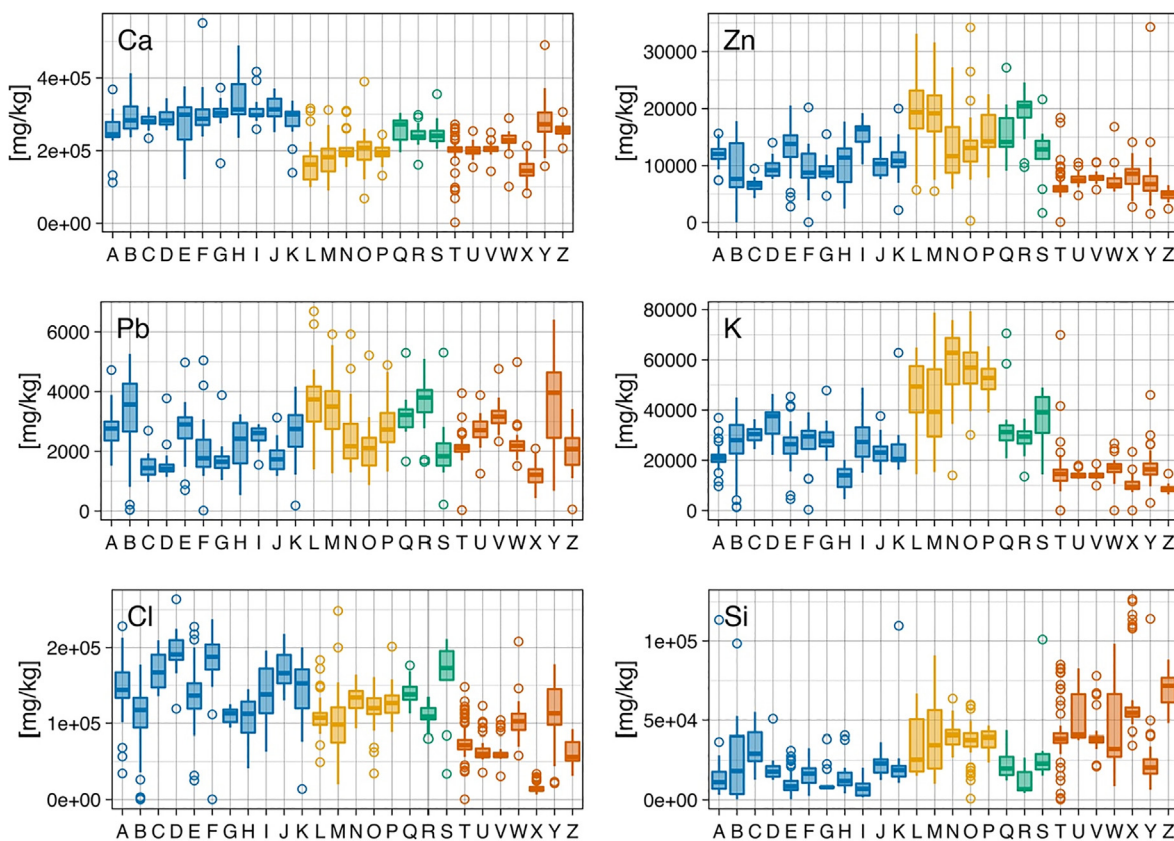
	Cu	Pb	Zn	Cd	Sb	Sn	Cr	Fe	Ca	Si	Al	S	Cl	Na	K
G-FA (97)	1200	11,000	28,000	650	530	1400	390	25,000	107,000	160,000	71,000	26,000	74,000	31,000	36,000
G-DS (97)	710	5400	15,000	300	790	890	180	12,000	230,000	69,000	26,000	15,000	180,000	17,000	23,000
G-FA (CHE)	2699	9704	45,160	280	2699	1444	569	20,344	174,263	78,150	38,325	54,493	104,215	46,951	46,218
G-APC (CHN)	990	2004	6370	121	-	-	121	-	-	-	-	-	-	-	-
FB-APC (CHN)	718	810	3105	42	-	-	42	-	-	-	-	-	-	-	-

variances of the elemental distributions for all the grouped incineration technologies. However, this assumption is not fulfilled for most elements, as shown in Fig. 5, from the homogeneity of variance test in Appendix E and from the tests presented in Appendix B. Therefore, the exact numerical values of  $\tau$  must be interpreted with care, but approximate conclusions can be drawn. Roughly, estimates show that the variation is twice as high within a grate incineration plant than between grate incineration plants – with a larger variation for DS residues than FA. In comparison, for FB-APC plants, the variation in residues are larger between plants than within the plants. Therefore, the FB-APC residues should be

well differentiated with respect to different FB technologies and flue gas cleaning systems and should be further investigated. This is substantiated with the different FB-APC clustering in the PCA (Fig. 1 A).

**4. Valorisation of MSWI APC residues**

Zn and Cu are considered the most valuable metals in APC residues. The Zn concentrations are higher in G-FA and G-DS (1 and 1.6 g/kg, respectively) than FB-APC residues (0.7 g/kg). The mean Cu concentration is higher in FB-APC residues (0.5 g/kg) than G-



**Fig. 5.** Zn, Pb, Cd, Cu, Ca and K elemental concentration of APC residues from different incineration plants A-Z. G-FA, G-DS and FB-APC are represented by yellow, blue and red colours, respectively. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

DS and G-FA (average of both 0.1 g/kg). For FB-APC residues, Cu should be the target metal for recovery. However, these described metal concentrations apply to Norwegian and Swedish APC residues as a result of their MSW management systems. The variation between Scandinavian, Chinese and Swiss MSWI APC residues illustrates that metal recovery processes must be adapted to geographical location and the location's holistic waste management system.

A high acid neutralization capacity of the APC residues results from the high Ca content (G-DS 1.5 higher mean than G-FA with 28.8 wt% compared to 18.7 wt%). This results in ideal properties for neutralizing spent industrial acid, as performed at the Langøya landfill. The high Ca concentrations may also allow the residues to be processed into suitable construction materials. However, the high Ca content can be challenging in metal extraction processes as high amounts of acid must be used to reach an acid extraction regime. The mean concentrations for Cl, Na, and K are about twice as high for grate incineration APC residues than FB residues, facilitating economic salt recovery.

The variation between different APC residues is important when considering the design of a material recovery process, especially if the process shall be centralized to handle several plants' residues. Larger variation should be expected from FB-APC than from G-DS, which should be expected higher than G-FA.

It is important to emphasize that the chemical concentrations only show the potential for material recovery. The concentration of organic pollutants, inorganic speciation and mineralogy in APC residues are not specified in this study yet are necessary for understanding separation processes in material recovery processes.

## 5. Conclusion

This article presents representative data on chemical composition of Swedish and Norwegian MSWI APC residues from 2006 to 2020. The APC residues are categorized as G-FA, G-DS and FB-APC residues, where the variation in chemical composition is presented and quantified. These variations must be taken into account when designing material recovery processes. This study clarifies several aspects concerning differences in chemical compositions for MSWI APC residues.

First, combustion technologies give rise to significantly different APC residues. G-FA and G-DS residues have higher concentrations of volatile elements compared to FB-APC residues, typically elements with low metal or metal oxide melting temperatures. The difference is evident for K, Na, Cd, Zn, Sb and Sn. In contrast, FB-APC residues have a higher concentration of Cu, Fe, Al, Si and Cr. This is due to lower combustion temperatures and more turbulent flows in the combustion chambers, which give a more even partitioning between bottom ash and APC residues. Large variations are observed in the FB-APC and are most likely coming from the different FB combustion technologies. The characteristics of different FB-APC residues should be further investigated and differentiated.

Second, various flue gas cleaning systems also give rise to significantly different APC chemical compositions. Differences between lime-based DS residues and FA from wet scrubber systems, both from grate incinerators, are quantified. Ca and Cl concentrations are higher for DS residues, while the other elemental concentrations are higher for FA. Further, chemical compositional variations are larger for G-DS than for G-FA due to more



considerable variation in DS designs than in dust filter designs for wet scrubber systems. Differences in DS and FA's chemical composition may indicate that Pb, S, Sb, Cd and Zn (in that order) tends to form stable gaseous species that can penetrate regular particle filters in wet scrubber systems.

Third, there is more considerable chemical compositional variation within MSWI plants than between MSWI plants. However, the variation between plants is significant. The variation is about twice within grate incineration plants than between grate incineration plants. The average relative standard deviation for elemental concentration within plants is 30%.

Fourth, metal recovery processes must be adapted to geographic variations, waste management systems and be robust considering chemical variation over time. The metal concentrations are significantly different for Scandinavian, Swiss and Chinese APC residues. Swiss FA is high in Zn, which is more beneficial for Zn recovery, while Chinese APC residues are low in valuable metals like Zn and Cu. The different metal concentrations of Scandinavian APC residues from 2006 to 2020 and FA reported by the IAWG (1997) illustrate the dynamic character of MSWI residues over time. Typical APC residue characteristics must be revised over time as MSW composition changes with time.

With an increased comprehension of the variation in chemical concentrations of MSWI APC residues, more insightful decisions regarding the development of material recovery processes may be taken. Hopefully, this might lead to more sustainable waste management practices for MSWI APC residues.

### Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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### Appendix A. Supplementary material

Supplementary data to this article can be found online at <https://doi.org/10.1016/j.wasman.2021.04.007>.

### References

Colangelo, F., Cioffi, R., Montagnaro, F., Santoro, L., 2012. Soluble salt removal from MSWI fly ash and its stabilization for safer disposal and recovery as road basement material. *Waste Manag.* 32, 1179–1185. <https://doi.org/10.1016/j.wasman.2011.12.013>.

Ebbinghaus, B.B., 1995. Thermodynamics of gas phase chromium species: the chromium chlorides, oxychlorides, fluorides, oxyfluorides, hydroxides, oxyhydroxides, mixed oxyfluorochlorohydroxides, and volatility calculations in waste incineration processes. *Combust. Flame* 101, 311–338. [https://doi.org/10.1016/0010-2180\(94\)00215-E](https://doi.org/10.1016/0010-2180(94)00215-E).

Elomaa, H., Seisko, S., Lehtola, J., Lundström, M., 2019. A study on selective leaching of heavy metals vs. iron from fly ash. *J. Mater. Cycles Waste Manag.* 21, 1004–1013. <https://doi.org/10.1007/s10163-019-00858-w>.

Falcoz, Q., Gauthier, D., Abanades, S., Flamant, G., Patisson, F., 2009. Kinetic Rate Laws of Cd, Pb, and Zn Vaporization during Municipal Solid Waste Incineration. *Environ. Sci. Technol.* 43, 2184–2189. <https://doi.org/10.1021/es803102x>.

Haberl, J., Koralewska, R., Schlumberger, S., Schuster, M., 2018. Quantification of main and trace metal components in the fly ash of waste-to-energy plants located in Germany and Switzerland: An overview and comparison of concentration fluctuations within and between several plants with particular focus on valuable metals. *Waste Manag.* 75, 361–371. <https://doi.org/10.1016/j.wasman.2018.02.015>.

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

Jurczyk, M., Mikus, M., Dziedzic, K., 2016a. Flue gas cleaning in municipal waste-to-energy plants - Part 1. *Infrastruktura Ekol. Teren. Wiej. nr IV/1*.

Jurczyk, M., Mikus, M., Dziedzic, K., 2016b. Flue gas cleaning in municipal Waste-to-Energy plants - Part 2. *Infrastruktura Ekol. Teren. Wiej. nr IV/2*.

Karlfeldt Fedje, K., Ekberg, C., Skarnemark, G., Pires, E., Steenari, B.-M., 2012. Initial studies of the recovery of Cu from MSWI fly ash leachates using solvent extraction. *Waste Manag. Res.* 30, 1072–1080. <https://doi.org/10.1177/0734242X12441385>.

Leckner, B., Lind, F., 2020. Combustion of municipal solid waste in fluidized bed or on grate - A comparison. *Waste Manag.* 109, 94–108. <https://doi.org/10.1016/j.wasman.2020.04.050>.

Life HaloSep [WWW Document], 2020. URL <https://lifehalosep.eu/>.

Liu, G.-Q., Itaya, Y., Yamazaki, R., Mori, S., Yamaguchi, M., Kondoh, M., 2001. Fundamental study of the behavior of chlorine during the combustion of single RDF. *Waste Manag.* 21, 427–433. [https://doi.org/10.1016/S0956-053X\(00\)00134-3](https://doi.org/10.1016/S0956-053X(00)00134-3).

Ma, W., Wenga, T., Frandsen, F.J., Yan, B., Chen, G., 2020. The fate of chlorine during MSW incineration: Vaporization, transformation, deposition, corrosion and remedies. *Prog. Energy Combust. Sci.* 76. <https://doi.org/10.1016/j.pecs.2019.100789> 100789.

Mian, M.M., Zeng, X., Nasry, A. Al N.B., Al-Hamadani, S.M.Z.F., 2017. Municipal solid waste management in China: a comparative analysis. *J. Mater. Cycles Waste Manag.* 19, 1127–1135. <https://doi.org/10.1007/s10163-016-0509-9>.

Moen, C., 2019. Christoffer Moen, Norcem. Private communication to Dag Eriksen, Feb.26, 2019.

Ohbuchi, A., Koike, Y., Nakamura, T., 2019. Quantitative phase analysis of fly ash of municipal solid waste by X-ray powder diffractometry/Rietveld refinement. *J. Mater. Cycles Waste Manag.* 21, 829–837. <https://doi.org/10.1007/s10163-019-00838-0>.

Östrem, S., 2019. Påverkansfaktorer på vätagasrelaterad reaktivitet hos CFB-flygaskor.

Quina, M.J., Bontempi, E., Bogush, A., Schlumberger, S., Weibel, G., Braga, R., Funari, V., Hyks, J., Rasmussen, E., Lederer, J., 2018. Technologies for the management of MSWI incineration ashes from gas cleaning: New perspectives on recovery of secondary raw materials and circular economy. *Sci. Total Environ.* 635, 526–542. <https://doi.org/10.1016/j.scitotenv.2018.04.150>.

Rio, S., Verwilghen, C., Ramarosan, J., Nzihou, A., Sharrock, P., 2007. Heavy metal vaporization and abatement during thermal treatment of modified wastes. *J. Hazard. Mater.* 148, 521–528. <https://doi.org/10.1016/j.jhazmat.2007.03.009>.

Ruth, L.A., 1998. Energy from municipal solid waste: a comparison with coal combustion technology. *ProgEnergyCombust Ci* 24, 545–564.

Tang, J., 2017. Removal and Recovery of Metals from Municipal Solid Waste Incineration Ashes by a Hydrometallurgical Process. Chalmers University of Technology.

Tang, J., Steenari, B.-M., 2015. Solvent extraction separation of copper and zinc from MSWI fly ash leachates. *Waste Manag.* 44, 147–154. <https://doi.org/10.1016/j.wasman.2015.07.028>.

The International Ash Working Group (IAWG), 1997. *Municipal solid waste incinerator residues, Studies in Environmental Science*. Elsevier, Amsterdam.

Wang, P., Hu, Y., Cheng, H., 2019. Municipal solid waste (MSW) incineration fly ash as an important source of heavy metal pollution in China. *Environ. Pollut.* 252, 461–475. <https://doi.org/10.1016/j.envpol.2019.04.082>.

Watanabe, N., Yamamoto, O., Sakai, M., Fukuyama, J., 2004. Combustible and incombustible speciation of Cl and S in various components of municipal solid waste. *Waste Manag.* 24, 623–632. <https://doi.org/10.1016/j.wasman.2004.03.003>.

Weibel, G., Eggenberger, U., Kulik, D.A., Hummel, W., Schlumberger, S., Klink, W., Fisch, M., Mäder, U.K., 2018. Extraction of heavy metals from MSWI fly ash using hydrochloric acid and sodium chloride solution. *Waste Manag.* 76, 457–471. <https://doi.org/10.1016/j.wasman.2018.03.022>.

Weibel, G., Eggenberger, U., Schlumberger, S., Mäder, U.K., 2017. Chemical associations and mobilization of heavy metals in fly ash from municipal solid waste incineration. *Waste Manag.* 62, 147–159. <https://doi.org/10.1016/j.wasman.2016.12.004>.