

Fate of heavy metals during municipal solid waste incineration in Shanghai

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Abstract

The transfer behavior of heavy metals during municipal solid waste (MSW) incineration was investigated based on 2-year field measurements in two large-scale incinerators in Shanghai. Great temporal and spatial diversification was observed. Most of Hg and Cd were evaporated and then removed by air pollution control (APC) system through condensation and adsorption processes, thus being enriched in the fine APC residues particles. Cr, Cu, and Ni were transferred into the APC residues mainly by entrainment, and distributed uniformly in the two residues flows, as well as in the ash particles with different sizes. Pb and Zn in the APC residues were from both entrainment and evaporation, resulting in the higher concentrations (two to four times) compared with the bottom ash. Arsenic was transported into the flue gas mainly by evaporation, however, its transfer coefficient was lower. Though the heavy metals contents in the APC residues were higher than that in bottom ash, more than 80% of As, Cr, Cu, and Ni, 74–94% of Zn, as well as 46–79% of Pb remained in the bottom ash, due to its high mass ratio (85–93%) in the residues. While 47–73% of Cd and 60–100% of Hg were transferred into the APC residues, respectively.

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Keywords: Municipal solid waste; Incineration; Heavy metals; Partitioning; Bottom ash; APC residues

1. Introduction

With the development of the social economy and improvement of living standards, increased amounts of municipal solid waste (MSW) are being produced in China. Incineration, as a waste treatment alternative benefiting volume reduction, thorough stabilization, sanitation, and energy generating, is playing a more and more important role in MSW management. A considerable number of new incineration plants are under construction or design. By the end of 2005, there were a total of 67 MSW incinerators with treatment capacity of 33,010 t/d in China, accounting for 13% of the MSW treatment capacity of the 471 treatment facilities [1]. Though MSW could be greatly reduced during incineration process, heavy metals contained in MSW are concentrated into the incineration byproducts, i.e., air pollution control (APC) residues, bottom ash, and exhaust gas, via physicochemical process.

The partitioning of heavy metals during incineration is influenced by many factors, which can be summarized into three categories [2–13]: (1) the occurrence and distribution of heavy metals in the incinerated waste, e.g., enriching whether in bulk waste or the inorganic/organic waste which is easily entrained into flue gas, physicochemical properties of the heavy metals influencing their evaporation or reaction dynamics kinetics; (2) the physicochemical condition that influences incineration, such as temperature, the composition of flue gas, chlorine in the waste, moisture content, etc.; (3) parameters influencing combustion kinetics, such as retention time, mixing conditions, type of grate, etc. In brief, higher temperature and high chlorine availability in gas favored evaporation of most metals, while oxidation/reduction, retention time, matrix composition, moisture content, etc., showed various effects on the transferring of specified metals. Sodium in the waste could increase the adsorption of heavy metals in the bottom ash due to its higher affinity for chlorine. Certain additives such as limestone, kaolinite, aluminum oxide, etc., were capable of reacting with these heavy metals and decreasing their volatilization during incineration.

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Most corresponding researches were conducted in bench-scale laboratory furnaces using model waste to investigate the factors influencing heavy metals transferring and partitioning during incineration [4–7,14–16], or by thermodynamics calculation to discuss and predict the chemical speciation and fate of heavy metals [10,17–20]. Only a few studies were carried out on the full-scale incinerators [8,21–24].

Jung et al. [8] observed that most Cd was intercepted in APC residues, while lithophilic metals (Fe, Cu, Cr, Al) and more volatile Pb and Zn remained mainly in the bottom ash. Yoo et al. [22] investigated 14 small-size incinerators and 4 boilers and found that volatile metals such as Cd, Pb, and Zn enriched in particulate matters in flue gas than in bottom ash. Chang and Ku [23] elucidated that more than 80% of Hg and As were transferred into the gas phase, while Cd, Cr, Cu, Ni, Pb, and Zn mainly partitioned in the bottom ash and APC residues. Similar results were obtained by Kuo et al. [24] on the partitioning behavior of Al, Cd, Cr, Cu, Fe, Pb, and Zn in a large MSW incinerator in Taiwan, with Cd primarily enriched in APC residues and other six metals concentrated in bottom ash. Based on material flow analysis on the bottom ash, boiler ash, gas cleaning residues, and exhaust gas, Belevi and Moench [21] suggested that Si, Fe, Co, Cr, Mn, Ni, P, Al, Ca, Mg, Na, Ba, Li, Ti, and K were transferred to the flue gas mainly by entrainment (without evaporation process), influenced by their occurrence and distribution in the input waste, while F, Cu, Mo, Pb, Sn, Zn, Br, Sb, C, S, Cl, As, Cd, and Hg were migrated to the gas by evaporation, determined by physical and chemical conditions as well as kinetics. The majority of these studies are based on once or twice sample analysis, which are hard to demonstrate the temporal and spatial diversification caused by the variation of MSW composition, incineration and air pollution control condition temporally and spatially.

In this study, partitioning and fate of heavy metals in the typical MSW of China – with higher water content and lower heat value – during full-scale incineration process were analyzed based on about 2 years of monitoring of heavy metals in the bottom ash and APC residues from two large-scale MSW incinerators in Shanghai. The incinerated MSW samples were simultaneously collected for 1 of the 2 years. Material flow analysis was used to elucidate the behaviors of heavy metals in the incinerators.

2. Materials and methods

2.1. The incineration residues samples

The water-quenched bottom ashes were sampled monthly from the ash pits of the two large-scale MSW incineration plants (incinerator J and incinerator P) in Shanghai, equipped with reciprocating grates. The treatment capacity of the two plants is approximately 1200 and 1500 t/d. The plants remove acid gas, heavy metals and dioxins, and particulates from the flue gas using lime slurry, activated carbon, bag filters, respectively. The resulting residues were collected from the semi-dry reactors and the bag filters, containing particles from the incineration chamber, reaction products, and some excess reactants, primarily $\text{Ca}(\text{OH})_2$.

From 2004 to 2006, a total of 28 bottom ash and APC residues samples were collected from incinerator J; while 13 bottom ash and APC residues samples were collected from incinerator P. The bottom ash samples were ground into less than $154\ \mu\text{m}$ before heavy metals analysis.

2.2. The incinerated MSW samples

The incinerated MSW samples were collected monthly from October 2004 to September 2005. For each sample, 100–150 kg of mixed MSW was collected from randomly selected locations in MSW pits of the incineration plants. The waste sample was then separated into nine fractions, i.e., plastics, paper, wood, textiles, metals, construction and demolition debris, glass, fruit waste, and vegetable waste. The total content of a certain heavy metal in the waste was calculated by summing up the amount of the metal in each fraction.

2.3. Mass balance of MSW flow during incineration

During October 2004 to September 2005, the total amounts of MSW transported into the two incinerators, the productions of leachate (pumped from the waste pit for in situ or ex situ treatment), bottom ash and APC residues were summed up respectively based on the daily operation data (by wet weight) provided by the owners. As the dosage of lime and activated carbon was relatively lower, contributing to less than 0.25% of the input mass, the mass balance was calculated as $\text{input (MSW)} = \text{output (leachate + bottom ash + APC residues + exhaust gas)}$. Then the mass of the exhaust gas could be obtained by subtraction, and the production ratios of the secondary pollutants could be obtained by dividing the quantity of the MSW by that of the corresponding pollutant.

2.4. Particle size separation

The APC residues sample was separated into six fractions (>154 , 74 – 154 , 54 – 74 , 38.5 – 54 , 30 – 38.5 , and $<30\ \mu\text{m}$) in terms of particle size by vibration sieve. Mineralogical and morphological analysis of the fractioned ashes was conducted using a D/max 2550 X-ray diffractometer (XRD, Rigaku Corporation, Japan) and a XL-30 environmental scanning electron microscopy (ESEM, Philips, the Netherlands). The soluble chlorides (Cl^-) and sulfates (SO_4^{2-}) were analyzed using AgNO_3 titration and BaSO_4 gravimetric method after extracting 25 g of the fractioned ash in distilled water at a liquid-to-solid ratio of 10:1.

2.5. Analysis of heavy metals

The total amounts of heavy metals in the ash and waste samples were analyzed by an AA320N atomic absorption spectrophotometer (AAS, Shanghai Analytical Instrument Overall Factory, China) (for Cd, Cr, Cu, Ni, Pb and Zn) and a XGY1012 atomic fluorescence spectrophotometer (AFS, Institute of Geophysical and Geochemical Exploration of the Chinese Academy of Sciences, China) (for As) after

Table 1
Mass flow of the waste in the MSW incinerators

Material flow	MSW	Leachate	Bottom ash	APC residues	Exhaust gas
Incinerator J (kg-wet)	1000	241	209	16	534
Incinerator P (kg-wet)	1000	101	210	27	662

HCl/HNO₃/HF/HClO₄ digestion. Mercury was analyzed using the AFS after HCl/HNO₃/H₂O₂ digestion.

2.6. Multivariate data analysis

Descriptive statistics and correlation analysis were applied to evaluate the analytical data using SPSS 13.0 for windows[®]. Independent samples *t*-test was conducted to elucidate the equality of means for the heavy metal contents in the incineration residues from the two incineration plants.

3. Results and discussion

3.1. Production ratio of the incineration residues

Table 1 shows the production of secondary pollutants during incineration, based on 1-year operational data. About 210 kg/t MSW (by wet weight) of bottom ash was generated in the two plants. The APC residues productions in the two incinerators were quite different, ranging from 16 to 27 kg/t MSW (by wet weight). The APC residues from incinerator J contained

more Ca(OH)₂ (data not shown), indicating that the higher APC residues production in incinerator P was not caused by lime dosage.

3.2. Occurrence and distribution of heavy metals in incineration residues

Descriptive analysis of heavy metals contents (by dry weight, the same as follows) in the incineration residues was conducted, as shown in Fig. 1. The data were within the reported [25] values for the heavy metals occurrence in the incineration residues. The wide range of metals contents indicated the great temporal variation, resulting from the strongly heterogeneous waste composition, the fluctuating incineration condition (such as temperature, excess air rate, perturbation of air current and heat current) and air pollution control parameters (e.g., lime and active carbon dosage).

The mean contents difference of heavy metals for the two incinerators was compared using independent samples *t*-test (Table 2).

The mean contents of As, Cd, Hg, Pb, and Zn in the residues from the two incinerators were statistically equal. While the contents of both Cu and Ni in the APC residues and bottom ash, as well as Cr in the bottom ash from incinerator P were significantly (at 0.05 significance level) higher than those from incinerator J, in spite of the similar occurrence of heavy metals in the incinerated MSW (Table 2) and the same incineration and APC process (mass burn equipped with grates, semi-dry APC system) adopted in the two incinerators. This

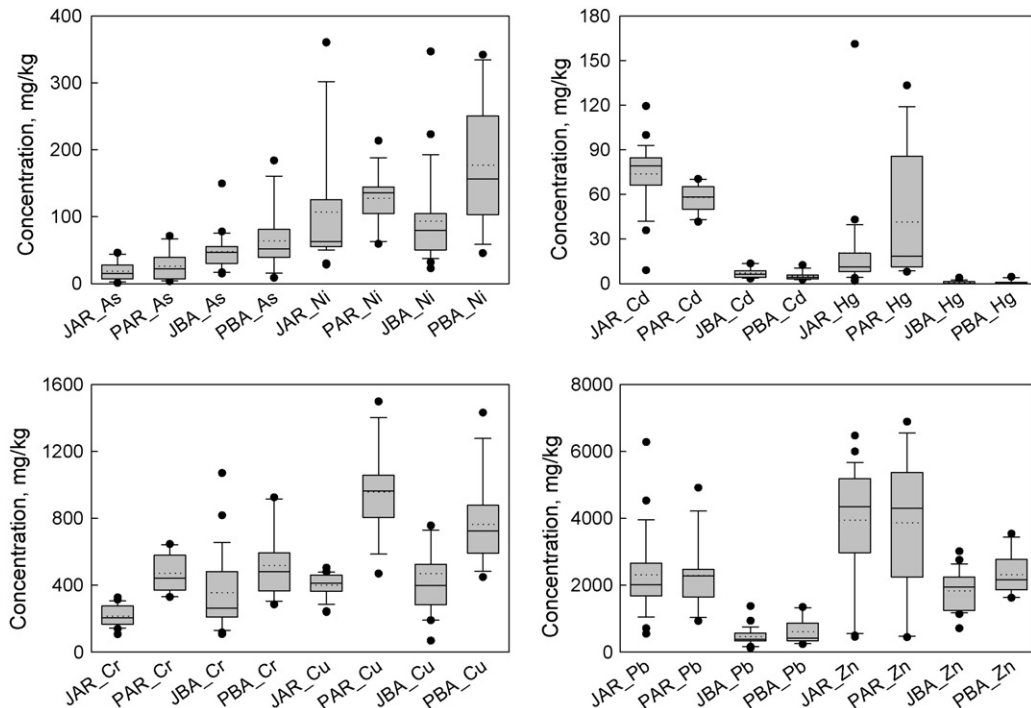


Fig. 1. Percentile contents of the heavy metals in the MSW incineration residues. Median, 10th, 25th, 75th, and 90th percentiles were plotted as horizontal solid lines of the boxes. The dashed line indicated the mean content value. Each outlier was shown as the dot. JAR and JBA, PAR and PBA denote the APC residues and bottom ash samples from incinerator J and P, respectively.

Table 2
Comparison of the mean contents of heavy metals in MSW incineration residues from the two incinerators

Heavy metals	Levene's test for equality of variances			t-Test for equality of means					
	Equal variances	F	Sig.	t	Sig. (two-tailed)	Mean diff.	Std. error diff.	95% confidence interval of the diff.	
								Lower	Upper
APC residues									
As	Equal variances not assumed	5.83	0.02	-1.89	0.08	-0.40	0.21	-0.86	0.05
Cd	Equal variances assumed	1.85	0.19	-0.83	0.42	-0.10	0.12	-0.35	0.15
Cr	Equal variances not assumed	24.6	0.00	-7.80	0.00	-10.0	1.28	-12.8	-7.23
Cu	Equal variances not assumed	12.0	0.00	-9.59	0.00	-19.8	2.06	-24.3	-15.3
Hg	Equal variances assumed	0.14	0.71	-0.69	0.50	-0.46	0.66	-1.84	0.92
Ni	Equal variances not assumed	8.05	0.01	-7.88	0.00	-2.59	0.33	-3.30	-1.88
Pb	Equal variances assumed	0.45	0.51	-1.42	0.17	-15.3	10.8	-37.7	7.09
Zn	Equal variances assumed	0.66	0.42	0.13	0.90	78.2	590	-1116	1272
Bottom ash									
As	Equal variances assumed	1.80	0.19	-0.50	0.62	-1.64	3.26	-8.40	5.12
Cd	Equal variances assumed	0.27	0.61	2.06	0.05	0.42	0.21	0.00	0.85
Cr	Equal variances assumed	0.00	0.99	-1.23	0.23	-22.7	18.5	-61.0	15.5
Cu	Equal variances assumed	0.09	0.76	-2.41	0.02	-57.1	23.6	-106	-8.04
Hg	Equal variances assumed	0.09	0.77	0.57	0.57	0.07	0.12	-0.18	0.31
Ni	Equal variances not assumed	5.67	0.03	-3.34	0.00	-19.5	5.84	-31.8	-7.18
Pb	Equal variances not assumed	5.27	0.03	-0.51	0.62	-13.2	25.8	-67.6	41.2
Zn	Equal variances assumed	0.02	0.89	-1.62	0.12	-79.4	49.1	-181	22.4
Incinerated MSW									
As	Equal variances assumed	4.24	0.05	1.69	0.11	26.6	15.8	-6.12	59.2
Cd	Equal variances assumed	3.89	0.06	-1.05	0.30	-0.68	0.65	-2.03	0.66
Cr	Equal variances assumed	1.07	0.31	0.01	0.99	0.42	42.7	-88.2	89.0
Cu	Equal variances not assumed	5.68	0.03	-1.68	0.11	-112	66.8	-254	30.7
Hg	Equal variances assumed	4.04	0.06	-1.24	0.23	-0.78	0.63	-2.08	0.52
Ni	Equal variances not assumed	16.4	0.00	-1.51	0.15	-15.4	10.2	-37.6	6.67
Pb	Equal variances assumed	3.88	0.06	-0.73	0.48	-26.8	36.8	-103	49.6
Zn	Equal variances assumed	0.66	0.43	-1.57	0.13	-69.6	44.3	-161	22.3

Note: sig.: significance; diff.: difference; Std.: standard deviation. Difference is significant at 0.05 level (two-tailed) when sig. <0.05.

could be explained by the difference of water and ash content in the two series of waste samples, which determined the final concentration ratio of the metals in the residues. The various incineration parameters, such as temperature, mixing condition, dosage of lime and active carbon, may also contribute to the diversity.

The contents of volatile heavy metals, Hg, Cd, Pb, and Zn in the APC residues, were obviously higher compared with bottom ash (Fig. 1), reaching to more than 40, 10, 4 and 2 times larger than that in the bottom ash, respectively. While the contaminant levels of other heavy metals, As, Cr, Cu, and Ni in the incineration residues were similar.

3.3. Mineralogy and morphology of the APC residues with different sizes

Figs. 2 and 3 show the trend of mineralogical and morphological variation of the APC residues with different particle sizes. Larger particles (>154 μm) were aggregated substances composed of melted silicates (such as quartz) which were transported from the furnace and reaction products (such as CaCO_3), while fine particles (<54 μm) with higher specific area contained more CaSO_4 , condensed salts (KCl, NaCl, etc.), and less silicates. Chemical analysis also proved the increase of chlorides and sul-

fates in the finer APC residues (Table 3). Therefore, volatile Hg, Cd, Pb, and Zn inclined to condense on the surface of these particles with chlorides, similar to the results of Wey et al. [26].

3.4. Fate of heavy metals during incineration

Transfer ratios of heavy metals to the APC residues (R_{iAR}) and bottom ash (R_{iBA}) were calculated by Eqs. (1) and (2). Where, AR and BA were the dry mass percentage of the APC residues and bottom ash in the incineration residues, respectively. C_{iAR} and C_{iBA} were the mass concentration of heavy metal i in the APC residues and bottom ash. The transfer ratios

Table 3
Concentration of dissolvable SO_4^{2-} and Cl^- in the ash particles

Ash particle (μm)	SO_4^{2-} (%)	Cl^- (%)
>154	0.56	3.16
74–154	0.58	2.77
54–74	0.68	3.68
38.5–54	0.71	4.78
30–38.5	0.81	7.02
<30	0.86	14.97

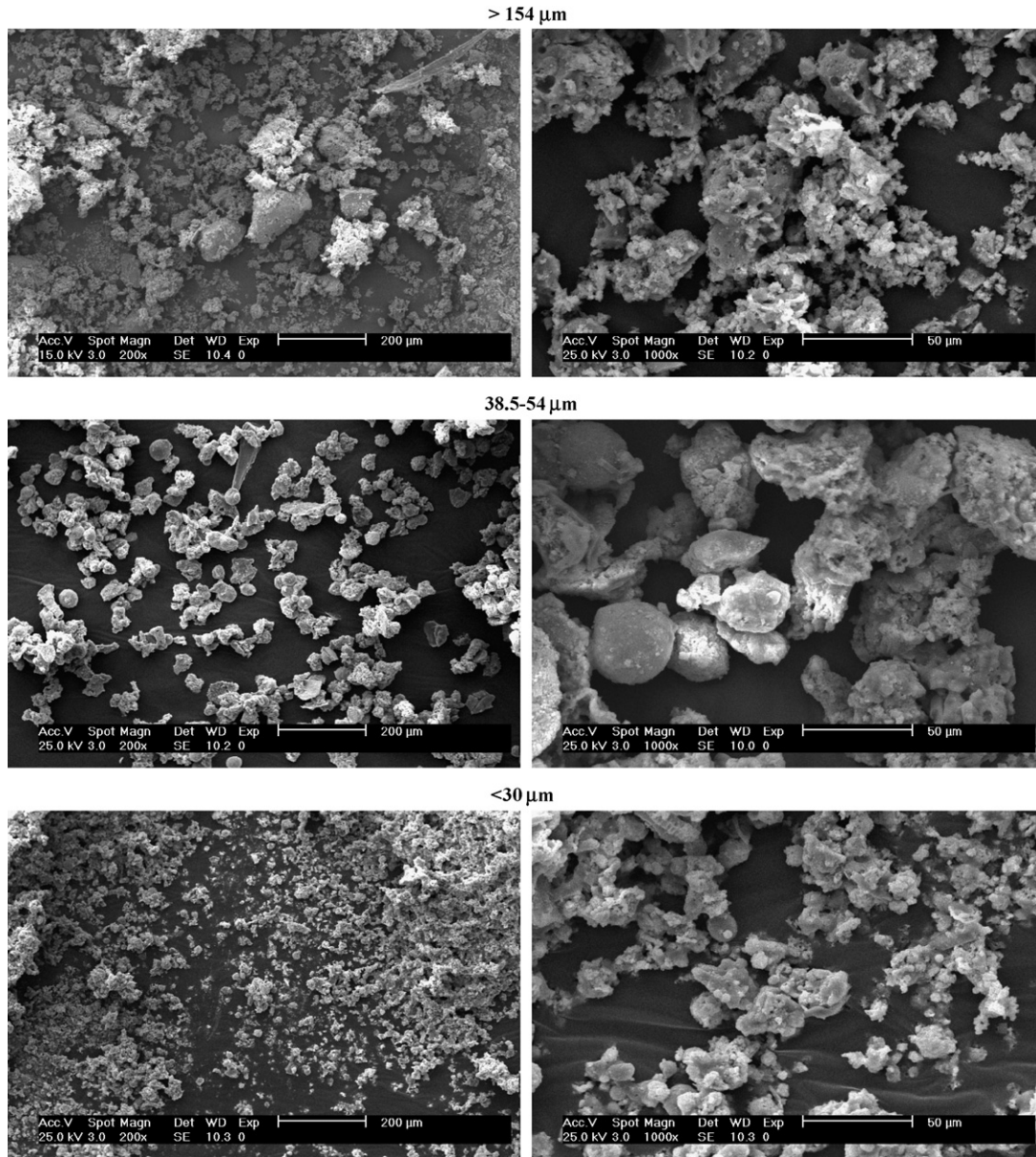


Fig. 2. ESEM photographs of the APC residues with different particle sizes.

of the APC residues were shown in Fig. 4:

$$R_{iAR} = \frac{AR(\%)C_{iAR}(\text{mg/kg})}{BA(\%)C_{iBA}(\text{mg/kg}) + AR(\%)C_{iAR}(\text{mg/kg})} \quad (1)$$

$$R_{iBA} = \frac{BA(\%)C_{iBA}(\text{mg/kg})}{BA(\%)C_{iBA}(\text{mg/kg}) + AR(\%)C_{iAR}(\text{mg/kg})} \quad (2)$$

Taking 10 and 90% percentile of contribution ratio as the range, As, Cr, Cu, and Ni showed lower transferability. After incineration, 89–99% of As, 85–97% of Cr, 80–93% of Cu and 83–95% of Ni still remained in the bottom ash. Volatile Cd and Hg were mainly in the APC residues, accounting to about 47–73% and 60–100%. Despite the high volatility of Pb and Zn, their transfer coefficients were relatively lower, only 21–54% and 6–26%.

Based on the thermodynamic calculation and experimental results [21,22], almost all of Hg and Cd in the MSW could volatilize and escape from the furnace with flue gas, which were then partly intercepted by the APC system via condensation, adsorption on fly ash or active carbon. Therefore, Hg and Cd were concentrated in the APC residues, determined mainly by their different thermodynamic characteristics and the physical and chemical conditions in the incinerators. Because condensation and adsorption occurs on the surface of particles, the content of Hg and Cd declined with the decrease of particle size of the APC residues, as shown in Fig. 5.

Lithophilic metals such as Cr, Cu, and Ni could hardly evaporate during incineration (Fig. 4), in accord with the field study results of Belevi and Moench [21]. However, Cu was different from the prediction by thermodynamic calculation, that most of

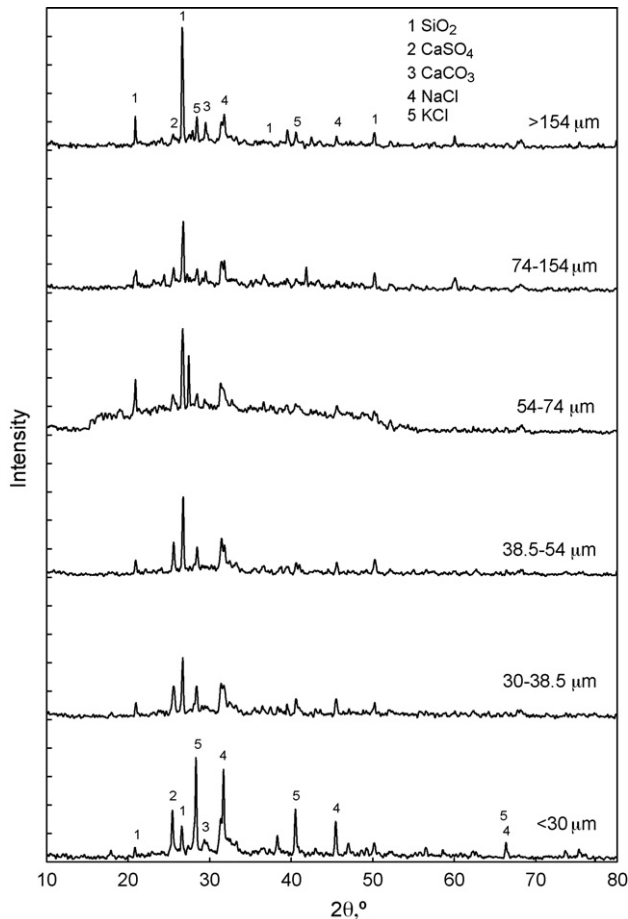


Fig. 3. XRD spectrum of the APC residues with different particle sizes.

Cu would be transferred to the flue gas as Cu_3Cl_3 and CuCl at $>700^\circ\text{C}$ [20]. This might result from the far lower affinity of Cu for chlorine compared with Na and Ca [12], and even with Pb and Cd [10]. Therefore, the three metals were transferred to the flue gas mainly by entrainment, which was proved by the significant bivariate correlation between their contents in the APC residues (Table 4). Occurrence of these elements in the bottom ash and APC residues was similar. It could be observed from Fig. 5 that they were distributed uniformly on ash particles

Table 4

Pearson correlation coefficients of the heavy metals contents in the APC residues

Pairs	As	Cd	Cr	Cu	Hg	Ni	Pb	Zn
As	1	-0.05	-0.03	-0.01	-0.11	-0.03	-0.50	0.11
Cd		1.00	-0.59	-0.58	-0.05	-0.63	0.16	0.04
Cr			1.00	0.80*	0.14	0.71*	-0.03	0.31
Cu				1.00	0.07	0.93*	0.05	0.23
Hg					1.00	0.15	-0.33	-0.41
Ni						1.00	-0.05	0.11
Pb							1.00	0.26
Zn								1.00

* Correlation is significant at the 0.01 level (two-tailed).

with different sizes, except the higher Cu content in ash particles sized $<30\ \mu\text{m}$, which was possibly due to both the entrainment and evaporation of Cu-enriched fine particles [27]. The transfer ratio of these metals was mainly determined by the parameters variation influencing entrainment in the furnace, such as waste composition, air feed rate, mixing on the grate, perturbation of air current and heat current.

Thermodynamic analysis showed that chlorides were the most stable forms for Pb and Zn over incineration temperature and they were expected to be transported to the gas [10,17]. The discrepancies between the calculated results and the field data could be explained by the difference in the amount of available Cl and reducing agent [20] as well as water content, etc. [18]. Both evaporation and entrainment determined the partitioning of Pb and Zn in the incineration residues. Therefore, although more Pb and Zn were found in the APC residues, the content difference of these elements between the APC residues and bottom ash was not as significant as that of Cd and Hg. Similar distribution behavior of Pb and Zn in the APC residues could be observed (Fig. 5), with increasing concentration in finer particles.

It had been reported [23,28] that As easily volatilized into the gas during incineration. However, a lower transfer coefficient was noticed in this study, similar to the field study result (76% retained in the bottom ash) of Belevi and Moench [21]. The majority of As was found to be contained in the inorganic part of the MSW [29] which was not melted under the incineration temperature (about 850°C). Besides, As could react with

Table 5

Comparison of the mean transfer coefficients of heavy metals in the APC residues from the two incinerators

Heavy metals	Levene's test for equality of variances			<i>t</i> -Test for equality of means					
	Equal variances	<i>F</i>	Sig.	<i>t</i>	Sig. (two-tailed)	Mean diff.	Std. error diff.	95% confidence interval of the diff.	
								Lower	Upper
As	Equal variances not assumed	4.42	0.05	-1.32	0.21	-6.37	4.81	-16.88	4.14
Cd	Equal variances assumed	0.18	0.68	-2.23	0.04	-9.94	4.45	-19.17	-0.70
Cr	Equal variances assumed	0.00	0.97	-5.67	0.00	-6.49	1.14	-8.86	-4.12
Cu	Equal variances assumed	1.36	0.26	-1.38	0.18	-4.30	3.13	-10.78	2.18
Hg	Equal variances not assumed	14.13	0.00	-2.13	0.05	-16.17	7.61	-32.48	0.13
Ni	Equal variances not assumed	5.05	0.03	-1.52	0.15	-3.05	2.01	-7.28	1.17
Pb	Equal variances assumed	0.65	0.43	-0.78	0.45	-4.42	5.70	-16.24	7.39
Zn	Equal variances assumed	0.19	0.67	-1.93	0.07	-5.57	2.88	-11.55	0.41

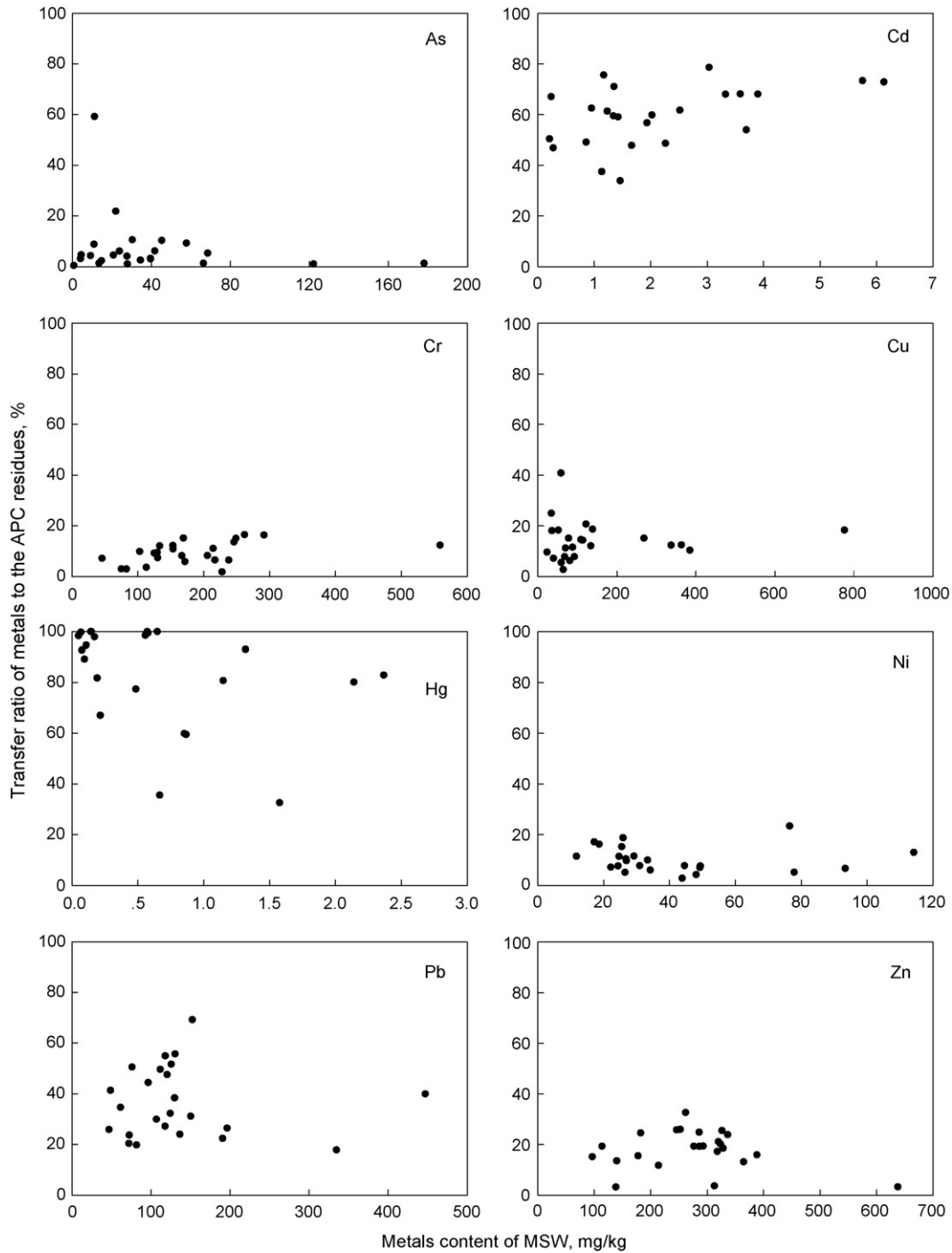


Fig. 4. Relationship between the transfer ratios of the heavy metals to the APC residues during incineration and the total amounts of heavy metals in incineration residues.

calcium oxide and form calcium arsenate, thus being retained in the bottom ash [6]. Fig. 5 demonstrated that the concentration of As increased with the decrease of particle size, which proved that As stayed in the APC residues was mainly from evaporation.

The partitioning of heavy metals in the incineration residues was not significantly correlated with the contents of these metals in the MSW (Fig. 4), which suggested that the incineration and the APC process condition may play a more important role.

The mean transfer coefficients of heavy metals to the APC residues from the two incinerators were compared (Table 5). Results suggested that both volatile metal (Cd) and lithophilic metal (Cr) could show different partitioning behaviors in the incinerators with similar MSW characteristics and incineration process. Temporal (as mentioned before) and spatial (incinerator-specific) variations were presented clearly by the field study.

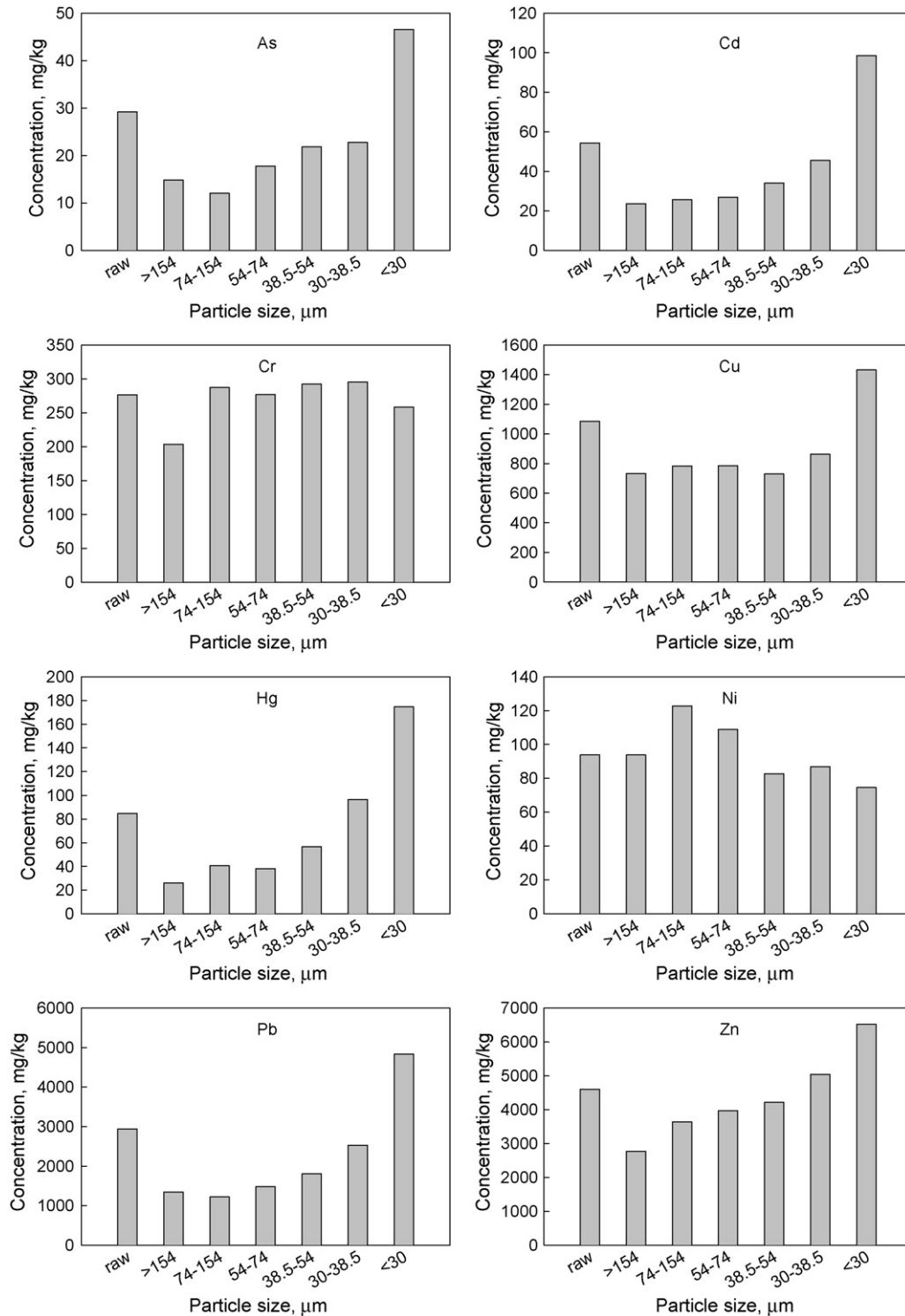


Fig. 5. Heavy metals content in the APC residues with different particle sizes.

4. Conclusions

According to the material flow analysis of the two MSW incinerators in Shanghai, about 210 kg of quenched bottom ash and 16–27 kg of APC residues were produced by incinerating 1 t of MSW (by wet weight). During incineration, most of Hg and Cd in the MSW were transferred into the APC residues by evap-

oration/condensation/adsorption process. As a result, contents of Hg and Cd in the APC residues were negatively correlated with the particle size of the residues, and were significantly higher than that in the bottom ash (40 and 10 times higher, respectively). Cr, Cu, and Ni were hard to be volatilized during incineration. They were transferred into the APC residues mainly by entrainment, and distributed uniformly on ash parti-

cles with different sizes. Therefore, the concentrations of these heavy metals in both bottom ash and APC residues were similar. Pb and Zn were transferred into the APC residues by both entrainment and evaporation, whose concentrations in the APC residues were relatively higher (two to four times) than that in the bottom ash. Evaporation was the main transfer process for As, however, because As was difficult to be released from the inorganic components which contained the majority of As in the MSW, the transfer coefficient of As from MSW to the APC residues was fairly lower.

Though the heavy metals concentrations in the APC residues were higher than those in bottom ash, most of the heavy metals were still retained in the bottom ash, due to the high composition (85–93%) of bottom ash in the incineration residues. The bottom ash contained more than 80% of As, Cr, Cu, and Ni, 74–94% of Zn, as well as 46–79% of Pb. While 47–73% of Cd and 60–100% of Hg were transferred into the APC residues, respectively.

Acknowledgements

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