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Characteristics of heavy metals on particles with different sizes from municipal solid waste incineration

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Abstract

Information on the concentration and size distribution of particles in the flue gas streams is essential for selecting and designing particle removal systems. Two municipal solid waste incinerators (MWIs) were selected for conducting flue gas sampling to determine the particulate distribution and heavy metals concentration on particles with different sizes by US EPA Method 5 sampling train and a cascade impactor. In addition, the characteristics of heavy metals contained on particles were investigated via isokinetic sampling of flue gas stream of air pollution control devices (APCDs). The experimental results indicated that average particulate matter (PM) concentrations at APCDs inlet were 2288.2±825.9 and 3069.2±810.0 mg/N m^3, while the concentrations of PM at stack were 1.51±0.20 and 14.81±4.52 mg/N m^3 in MWI-A and MWI-B, respectively. The differential mass size distribution of PM and differential elemental size distribution of Zn, Pb, and Cu in front of APCDs were of bimodal forms. Results indicate that Zn>Pb>Cu in order of mass concentration in each stage. The fine particles represent approximately 70% and the coarse particles account for the rest 30% of total particulate matters collected on eight stages for both incinerators. Zn, Pb and Cu on fine particles account for approximately 80% and those on the coarse particles are less than 20% of the total heavy metals collected on eight stages of the cascade impactor for both incinerators.

Keywords: Municipal solid waste incinerator; Particles; Isokinetics sampling; Heavy metals

1. Introduction

In Taiwan, space for sanitary landfill is expensive and extremely difficult to find due to the relatively high population density. As a result, Taiwan government is engaged in building more than 20 large scale waste-to-energy facilities to treat more than 90% of the municipal

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solid wastes (MSW) generated in the island during the next 3 years. Although the mass and volume of wastes can be effectively reduced by incineration, heavy metals may be present in the form of fly ash or vapor in flue gas in the meantime. Potential emissions of heavy metals from these municipal solid waste incinerators (MWIs) have caused much public concerns. Emission characteristics of heavy metals from MSW incinerators are affected by various operating parameters including the concentration of metals in the waste feed, composition of the gas stream, combustion temperatures, and the performance of the air pollution control devices (APCD) [1].

Pacyna estimated that in Europe, 3.0, 1.7, 0.7 and 7.0% of the emissions of Cd, Cu, Pb and Zn, respectively, originated from refuse incineration [2]. Morseli indicated that Cr and Mn were mainly retained in the slag (70%), while Pb, Zn and As were equally partitioned between fly ash and slag. Nevertheless, the metals mentioned above only have a small fraction (less than 5%) in flue gas [3]. Due to the relatively high vapor pressure, Hg is predominant as gas phase in flue gas. Vogg indicated that more than 66% mercury in the flue gas would penetrate the APCDs and be released into the atmosphere in a typical MSW incinerator [4]. Brunner and Monch indicated that in the MSW incineration process heavy metals might be transformed to mercuric chlorides and had higher vapor pressures than oxidized or elemental metals [5]. Therefore, high concentration of chlorine in waste materials would increase the emission of heavy metals from MWIs. Generally speaking, different types of APCDs have different levels of removal efficiencies for heavy metals in MWIs. Dhargalkar and Goldbach indicated that flue gas temperature and particle size were major factors affecting the removal and emission of heavy metals from gas streams. The emission information listed in AP-42 indicates that except for Hg, the removal efficiencies of heavy metals are at least 90% with different types of APCDs including ESP, SD/ESP, DSI/FF and SD/FF [6]. Dhargalkar and Goldbach indicated that the concentration of heavy metal emissions ranged from less than 0.00037 mg/N m^3 (12% CO_2) each for arsenic, beryllium and selenium to 0.3360 mg/N m^3 for nickel at fabric filter outlet of MWI. As far as the mercury is concerned, the removal efficiency is typically less than 30% with the corresponding emission level of 0.20 mg/N m^3 [1].

In the waste incineration process, particles can be formed via two possible ways: (1) homogeneous nucleation of metal (in either chloride or oxide form) vapor, followed by the growth of these nuclei via agglomeration and heterogeneous condensation. This process results in the formation of small particles, typically with the size smaller than 1 μm. (2) Transformation of ash forming material in the wastes (including mineral particles and inorganic salts) on the incomplete combustion particles to form fly ash. This process generates large particles, typically with the size larger than 1 μm. Two mechanisms for particulate matter formation occur at the same time, and the rates of reactions are related to the boiling point of heavy metals, the temperature, chlorine and sulfur contents of MSW. Kauppinen found that mass size distributions of particles were bimodal in the refuse incinerator equipped with a cyclone [7]. Two modes including fine and coarse mode were observed in size distribution. The geometric aerodynamic mean diameter of the fine mode varied between 0.1 and 0.2 μm, while the coarse mode mean diameter ranged from 6 to 10 μm. Mg, Al, Cl, Ti and Fe were found only in coarse particles. Elemental size distribution of Na, K, Zn, Cd, Pb and S were also of bimodal form. Furthermore, over 90% of the particle phase of S and 62–77% of Cd, respectively, were found in the fine particles [7].

Information on particulate matter (PM) concentration and size distribution upstream of APCDs is essential for planning and designing air pollution control devices for new MWIs and for assessing the removal efficiency of existing APCDs. Various kinds of sampling instruments methods have been developed to characterize the PM emissions from stationary combustion processes, such as konimeters, cascade impactor, impingers, precipitators, and particle counting and sizing instrument [8]. Some imperfection and sampling and analysis errors of these instruments were recognized while the methods were first developed. The common encountered problems of PM size separation instrument include particles rebound, re-entrainment and disaggregation during sampling [8]. The sampling times of different instruments varies from seconds to several hours. Moreover, the collection efficiency of different instrument also varies substantially. As a result, a comparison of the concentrations measured by various instruments is difficult. The differences in the measured concentrations with different instruments commonly lay in the range of ±100% [8]. Hence, no single conversion factor is available for comparing particle measurements made by two different instruments [8]. The importance of isokinetic sampling in the measurement of aerosol has been recognized since 1960s for obtaining representative samples [9].

2. Experimental

2.1. Location

Two large-scale municipal solid waste incinerators (designated as MWI-A and MWI-B) in northern Taiwan were selected for conducting flue gas sampling to determine the characteristics of particle size distribution and heavy metal emissions. Both incinerators are of mass-burn type and are equipped with different types of APCDs. MWI-A is equipped with a cyclone, dry lime scrubbing system with fabric filters while MWI-B is installed with an electrostatic precipitator (ESP) followed by wet scrubbers for removing air contaminants from flue gas streams. Both MWIs were operated at the temperature of 850–1050°C. The gas temperature and pressure drop of the fabric filters in MWI-A were controlled at 150±10°C and 200±20 mmH_2O, respectively. As for the MWI-B, the inlet gas temperature of the BSP was controlled at 240±20°C to prevent its corrosion and the pressure drop across the ESP was around 30 mmH_2O. The proximate analysis of incoming MSW in northern Taiwan indicated that it contained 44.5–49.6% moisture, 13.6–15.3% ash and 36.8–41.9% combustible. Furthermore, results of elemental analysis of the combustible indicated that it contained 21.0–23.6% carbon, 2.8–3.2% hydrogen, 12.4–14.0% oxygen, 0.2–0.4% nitrogen and 0.2–1.1% chlorine. The sampling points in front of APCDs were selected to characterize the PM and heavy metals concentrations leaving the incinerating chamber while the sampling points of stacks were selected to evaluate the emission characteristics as well as the removal efficiencies of APCDs.

2.2. Instrument

A stack sampling system equipped with cascade impactor complying with the US. EPA Method 5 was used as the sampling train. Four impingers are included in the

sampling train for Method 5. The first and second impingers contain distilled water to remove most of the moisture and corrosive matter to prevent the dry gas meter from damage. The third impinger is empty to remove moisture. In addition, the fourth impinger containing silica gel is connected to ensure the flue gas is thoroughly dried before it leaves the impingers. Since a significant portion of heavy metals in flue gas is bound to PM, isokinetic sampling has to be conducted to collect a representative sample. In this study, cascade impactor is selected for determining the particle size distribution at APCD inlets and stack. The cascade impactor consists of ten crosshair gaskets, ten inconel spacers, and ten plates which are numbered as 0, 1, 2, 3, 4, 5, 6, 7, 8 and F stage from top to bottom. Plate 0 is an orifice stage to get uniform velocity distribution in the impactor Plate F is a holder to backup filter to collect the particles penetrating the eight stages. The pre-separator of cascade impactor is used to eliminate the particles with diameter larger than 15 μm to avoid the overloading of the impactor. The impactor is tightly screwed in the head of the probe to prevent from falling during sampling period. The principle of cascade impactor is based on the different impaction parameter of the aerosols with different sizes between a series of parallel plates to distinguish particle sizes from a group of aerosols. The size distribution of particulate matter can be characterized by the effective cutoff diameter of particle (μm), D_{50}, which is defined as [10]

$$D_{50} = \sqrt{\frac{18\mu D_c \psi}{C_o \rho_p V_o}}$$

where μ is the viscosity of air (poise), D is the diameter of the round jet (cm),ψ is the dimensionless inertial impaction parameter; C_o is the Cunningham slip correction factor $(1+0.16\times10^{-4}/D_p)$ for normal temperature and pressure, ρ_p is the density of particles (g/cm^3), and V_o is the velocity of aerosol jet (cm/sec). The aerodynamic cutoff diameters of eight stages are 10.9, 8.2, 6.8, 4.6, 3.2, 2.0, 1.0 and 0.6 μm, respectively.

The mass concentrations of particulate matter and heavy metals in gas streams were evaluated via isokinetic sampling conducted on seasonal basis for both incinerators during one-and-half year study period. Sampling time at upstream of APCD and stack typically lasted for 20 min and 4 h, and sampling volumes are approximately 0.1 and 4 N m^3, respectively.

2.3. Analytical techniques

After the sampling is completed, the multistage filters are conditioned and weighed by Mettler M3 balance (0.01 mg precision) to obtain particulate mass on each stage. The size-fractionated particle samples were then digested with US EPA Method SW 846 3050A for heavy metal analysis. The accuracy of these measurement was checked by analyzing CRM0 19-050 standard ash certified by US EPA and the results were in good agreement with those specified. Zn, Pb and Cu concentrations were measured by atomic absorption spectrophotometry (AAS) technique with Varian, Model 400 Atomic Absorption Instrument.

Table 1
Concentrations and removal efficiencies of particulate matter in MWI-A and MWI-B (based on 10% O_2)

	MWI-A			MWI-B		
	PM at APCD inlet (mg/N m^3)	PM in stack (mg/N m^3)	Removal efficiency (%)	PM at APCD inlet (mg/N m^3)	PM in stack (mg/N m^3)	Removal efficiency (%)
Run 1	3944.2	1.47	99.96	3799.7	18.37	99.52
Run 2	1804.4	1.19	99.93	3927.9	17.99	99.54
Run 3	2232.2	1.72	99.92	2197.7	8.17	99.63
Run 4	1844.3	1.61	99.91	1982.7	10.30	99.48
Run 5	1891.4	1.38	99.93	3228.4	15.36	99.52
Run 6	2012.5	1.66	99.92	3278.7	18.66	99.43
Average	2288.2	1.51	99.93	3069.2	14.81	99.52
S.D.	825.9	0.20	0.02	810.0	4.52	0.07

3. Results and discussion

To discuss the relationships of PM with different sizes and heavy metal concentration, a cascade impactor was used to evaluate particle distributions in front of APCDs and sampling the PM at stack to evaluate the removal efficiencies of PM. Table 1 shows the concentrations and removal efficiencies of PM in flue gas of MWI-A and MWI-B. Results indicated that average PM concentrations in front of APCDs were 2288.2±825.9 and 3069.2±810.0 mg/N m^3 that concentrations at stack were 1.51±0.20 and 14.81±4.52 mg/N m^3 (based on 10% O_2) in MWI-A and MWI-B, respectively. The typical PM concentration used for designing the APCDs of MWI is 8000 mg/N m^3 (based on 12% CO_2), which is significantly higher than the values obtained in this study [11] RSD (%) of concentrations in front of APCDs are up to 36.09 and 26.39% for MWI-A and MWI-B, respectively. The relatively large RSD observed in front of APCDs is possibly attributed to the regular cleaning of economizers with high pressure air for increasing the heat exchange rate. The cleaning is conducted typically 3 or 4 times per day. The purge air can effectively blow off the fly ash from the boiler surface into the gas stream and results in a sudden increase of particle concentration. The removal efficiencies of PM at MWI-A and MWI-B are 99.93 and 99.52%, respectively. The removal efficiency of baghouse is significantly higher than that of ESP possibly due to the variation of particle's resistivity. Particle's resistivity is extremely

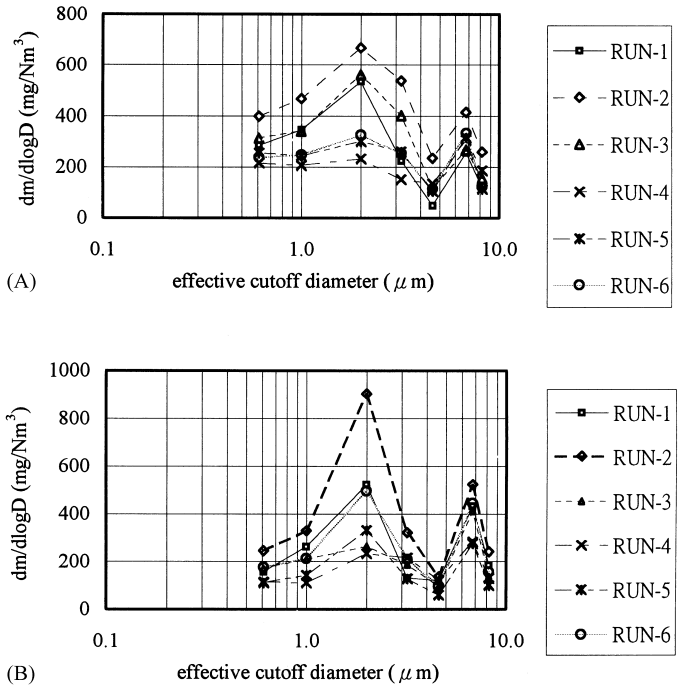

Fig. 1. (A) Particle mass concentration vs. cutoff diameter in MWI-A; (B) particle mass concentration vs. cutoff diameter in MWI-B.

Table 2
Total concentrations and distribution of heavy metals in front of APCDs between fine and coarse particles (based on 10% O_2)

	MWI-A			MWI-B		
	Total concentration ($mg/N\,m^3$)	Fine particles (%)	Coarse particles (%)	Total concentration ($mg/N\,m^3$)	Fine particles (%)	Coarse particles (%)
Zn	15.5–260	75–92	8–25	12.7–34.2	79–88	12–21
Pb	6.3–26.0	80–96	4–20	3.8–22.4	81–92	8–19
Cu	1.1–1.6	75–94	6–25	0.6–2.2	78–89	11–22

important because it strongly affects particle collection efficiency achieved with ESP. However, the resistivity of fly ash generated by MSW incineration varies widely and may not be within the optimal operation range of ESP due to the complexity of feeding wastes.

Particle mass concentrations versus aerodynamic cutoff diameters collected at APCD inlet for both incinerators are presented in Fig. 1A and B. Two modes are observed for the distribution and the trends of fine mode and coarse mode are quite similar for both MWIs. Results indicate that the concentrations of fine mode generated by vapor condensation are higher than that of coarse mode formed by mechanical forces. Table 2 indicates that the total collected PM consists of approximately 70% fines and 30% coarses or so of total

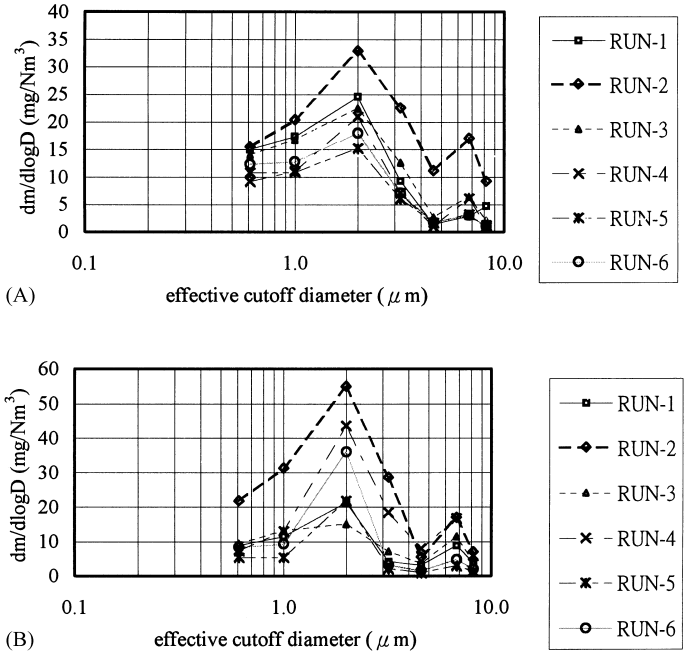

Fig. 2. (A) Concentration of Zn on different particle size sampled from MWI-A; (B) concentration of Zn on different particle size sampled from MWI-B.

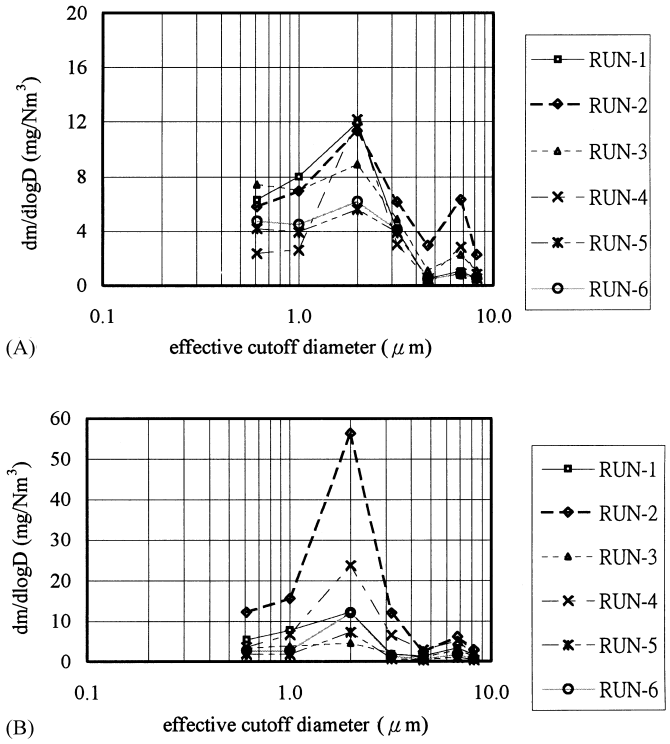

Fig. 3. (A) Concentration of Pb on different particle size sampled from MWI-A; (B) concentration of Pb on different particle size sampled from MWI-B.

PM collected on eight stages in both incinerators. Figs. 2–4 demonstrate the respective concentration of Zn, Pb and Cu contained in particles versus aerodynamic cutoff diameter of particles sampled at APCD inlets of both incinerators. Results indicate that the mass concentrations of Zn, Pb and Cu are of bimodal form and the trends agree with that of Kauppinen in 1990. The fine mode, located around 2 μm is formed partly by the agglomeration of fine particles and partly by the conversion of gases and vapors to particles. The fine particles are collected between the eighth and fifth stage filter (0.6–3.2 m). Based on the aerosol theory, the formation of fine mode at incineration system is mostly generated by the condensation of heavy metal vapors. The aerodynamic cutoff diameter of the coarse mode is 6.8 μm. The coarse particles are collected from the fourth to first stages of the impactor (4.6–10.9 μm). The elemental differentiation distributions indicate that the heavy metals are more enriched on fine mode than on coarse mode. Zn, Pb and Cu on fine particles account for approximately 80% of the total mass and the rest 20% are found on coarse particles for both incinerators. Major amounts of heavy metals on the fine particles are formed mainly by condensation and aggregation due to the higher surface area of fine particles. Therefore, major part of Zn, Pb and Cu exists on the particles with the cutoff diameter less than 3.2 μm. Fig. 5 shows that Zn, Pb and Cu concentrations on the particles with different sizes have similar trends and the order of mass concentration on particles is Zn>Pb>Cu for

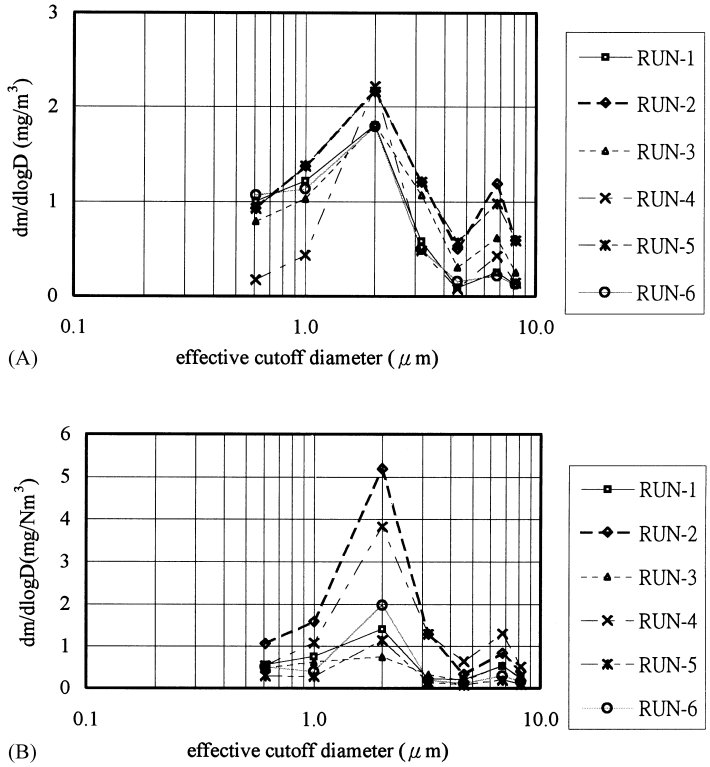

Fig. 4. (A) Concentration of Cu on different particle size sampled from MWI-A; (B) concentration of Cu on different particle size sampled from MWI-B.

both incinerators. The distributions of particulate matter and heavy metal concentration at APCD inlet in MWI-B are similar to that in MWI-A.

Because fine particles penetrate deeper into the lung, the human health is significantly influenced by the synergism of fine particles and heavy metals associated with them despite of the relatively small percentages of fine particles in total particulate matter (including the particles with diameter larger than 10.9 μm). Correspondingly, research and development of advanced technology for effective removal of fine particles from MWIs is essential for reducing the risks caused by fine particles and heavy metals.

4. Conclusions

The average concentrations of PM in front of APCDs were 2288.2±825.9 and 3069.2± 810.0 mg/N m^3, while the concentrations of PM on stack were 1.51±0.20 and 14.81± 4.52 mg/N m^3 in MWI-A and MWI-B, respectively. The removal efficiencies of MWI-A and MWI-B for particulate matter are 99.93 and 99.52%, respectively. The removal efficiency of baghouse for particulate matter is considerably higher than that of electrostatic precipitator.

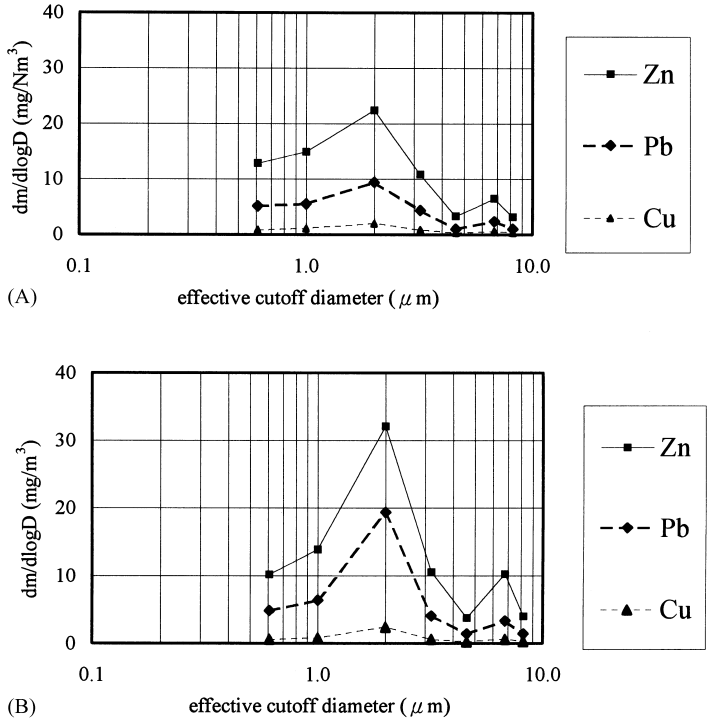

Fig. 5. (A) Concentration of Zn, Pb and Cu on different particle size sampled from MWI-A; (B) concentration of Zn, Pb and Cu on different particle size sampled from MWI-B.

The distributions of PM, Zn, Pb and Cu in front of APCDs are bimodal (i.e. fine and coarse modes). For both MWI-A and MWI-B, the results demonstrate that Zn>Pb>Cu in order of the mass concentration. The size fractional particles and heavy metals concentrations at APCD inlet in MWI-B are similar to that of MWI-A despite the differences in geographic location. The size distributions of Zn, Pb and Cu from incineration chambers have similar trends. The fine particles account for approximately 70% of total PM collected on eight stages in MWI-A and MWI-B.

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