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Particle Size Distribution of PM-10 and Heavy Metal Emission with Different Temperature and HCl Concentrations from Incinerators

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Abstract–Emission characteristics of particulate matter and heavy metals from 12 small waste incinerators, whose capacity ranged from 25 to 200 kg/h of waste, were investigated to determine the factors affecting the particulate matter generation and growth mechanisms. The ratio of fine particles to coarse particles increased with the flue gas temperature. Particulate matter showed bimodal forms in particle size distributions. The finer particle mode in particle size distribution shifted toward the coarser particle mode with a decrease in flue gas temperature. Experimental results were in agreement with coagulation theory: It is thought that the coarser particles were mechanically generated and the finer particles were generated by gas-to-particle conversion mechanisms such as nucleation, condensation, and coagulation. Heavy metal enrichment in finer particulate matter was also observed and related to particle formation and growth from vaporized metals. Emission of all heavy metals except zinc was affected by hydrogen chloride concentrations, while some metal emissions such as manganese, chromium, and copper were not varied with flue gas temperature.

Key words: Particle Size Distribution, Heavy Metals, Incinerators, Particle Growth, Nucleation

INTRODUCTION

Many small incinerators (capacities under 200 kg/h waste combustion) have been installed and have been in operation for the last two decades in Korea. Because of difficulties in their management with proper regulatory compliance due to stricter standards of air pollutant limits, the government has established a strategy not to permit any more construction of such small incinerators or to let them install an appropriate air pollution control device (APCD) even though economically infeasible.

In courses of investigating emission characteristics of particulate matters (PMs), particle size distribution (PSD) and heavy metal enrichment in particles have been studied for better understanding of their emission and control. Many research works have investigated emission characteristics of hazardous air pollutants from fuel combustion and waste incinerators. Particularly, PM contains many hazardous heavy metals and PM-2.5 (PM less than 2.5 micrometers in aerodynamic diameter) or PM-10 (PM less than 10 micrometers in aerodynamic diameter) are easily deposited in the human bronchus. In the waste incineration process, PMs 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 of a size smaller than 1 micrometer; [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 of a size larger than 1 micrometer [Chang et al., 2000].

From the two different formations of particles, less and bigger than 1-3 micrometers, some investigators have classified them into the finer, as generated by nucleation/vaporization/condensation, and the coarser, as mechanically generated [US EPA, 2001; Linak et al., 2000]. There are some extended and reviewed papers for metals partitioning and their formation mechanism and control in the combustion process based on numerous data and equilibrium calculation results by Linak and Wendt [Linak and Wendt, 1993; Linak, 2000].

Most of the studies concerning PSD and heavy metal emissions have been performed at either a well-arranged experimental condition or a single large-scale incinerator. Therefore, such finer and coarser particle formation in real small incinerators, which have been widely utilized in the Korea, could be difficult to correlate with and to understand their emission characteristics. In this study, PSD and emission concentration of PMs such as total particulate matter (TPM) and PM-10 from 12 small size incinerators (capacities between 25-200 kg/h waste combustion) were investigated to observe the fine particle formation by metal nucleation/coagulation/condensation. A simple coagulation model to study particulate growth after the nucleation of metals has been applied and compared with the experimentally obtained PSD from incinerators. Additionally, heavy metal emission has also been introduced with real field measurement data while most of the research for such fine particles and heavy metal emission was conducted in experimental set-ups under precisely controllable and manageable conditions.

EXPERIMENTS

1. Tested Facilities

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‡This paper is dedicated to Professor Dong Sup Doh on the occasion of his retirement from Korea University.

Table 1. Identifications, capacities and types of tested incinerators

ID*	Capacity (kg/hr)	Fuel (composition in %)	Incinerator type
MW-1	50	Paper (30), synthetic resin (30), wood (30), textile (10)	Semi Batch
MW-2	150	Paper (55), synthetic resin (18), wood (17), cotton+textile (10)	Semi Batch
MW-3	190	Paper (69), synthetic resin (6), rubber (4), wood (17), textile (4)	Semi Batch
IW-1	30	Paper (52), synthetic resin (35), wood (8), textile (5)	Batch
IW-2	30	Paper (30), synthetic resin (20), wood (48), textile (2)	Batch
IW-3	30	Paper (35), synthetic resin (65)	Batch
IW-4	50	Paper (40), synthetic resin (30), wood (20), textile (10)	Batch
IW-5	80	Paper (22), synthetic resin (40), wood (15), textile (5), styrofoam (2), rubber (5), redder (3), others (8)	Semi Batch
IW-6	95	Paper (sand paper, 42), wood (50), textile (6), vinyl (2)	Semi Batch
IW-7	95	Vinyl (81), paper (15), wood (4)	Semi Batch
IW-8	95	Synthetic resin (47), textile (17), wood (11), paper (25)	Batch
IW-9	150	Paper (coated, 41), textile (49), vinyl (10)	Semi Batch

*MW: Municipal waste incinerator
IW: Non hazardous industrial waste incinerator

The example facilities were chosen randomly from among the installed facilities. All of the small size incinerators (MW: municipal waste and IW: industrial waste) have simple air pollution control devices (APCDs) such as a cyclone. Table 1 shows a summary of the tested incinerators in this study. The sampling location was the APCD (cyclone) outlet because most of the small incinerators were unavailable for sampling at the APCD inlet due to the high temperature of flue gas and the short length of duct.

2. Experimental Methods

The sampling method for TPM used in this experiment was based on the iso-kinetic sampling method (Korea Standard Method for Air Pollution [NIER, 1999]), which is similar to the US EPA method 5 [US EPA, 2000]. The cascade impactor (Anderson Instrument Co. Ltd., Mark III Stack Sampler) was used for PM-10, PM-2.5 sampling, and particle size distribution analysis. It was adapted for EPA method 201A-Determination of PM-10 Emission [US EPA, 1997]. The size distribution was calculated by WinCIDRS software (WinCIDRS, Ver. 4.0 Operations and Data Analysis System for Internal Particle Sizing Device).

The US EPA test method for evaluating solid waste, SW-846 3050B [US EPA, 1986], was used for digesting the filter collected dusts for heavy metal and inorganic matter analysis, and their concentrations were analyzed by using ICP/MS (Varian Co. Ltd., Ultra mass 700).

RESULT AND DISCUSSION

1. Emission Concentrations and Ratios of PMs

The emission concentrations and ratios of PMs such as TPM, PM-10, PM-2.5 and PM-1 (PM less than 1 micrometer in aerodynamic diameter) are shown in Table 2. Among the small size incinerators, even TPM concentration was scattered with a standard

Table 2. Concentrations and ratios of particulate matter from incinerators

ID	Concentration (mg/Sm^3)*				PM-10/ TPM (%)	PM-2.5/ PM-10 (%)	PM-1/ PM-10 (%)	Flue gas temp. °C
	TPM	PM-10	PM-2.5	PM-1				
MW-1	538.1	161.0	158.0	146.0	30	98	91	432
MW-2	180.1	34.0	29.1	21.8	19	86	64	133
MW-3	419.9	43.5	26.2	15.6	10	60	36	122
IW-1	126.3	82.2	78.0	74.9	65	95	91	633
IW-2	185.8	85.2	79.3	73.5	46	93	86	461
IW-3	249.0	96.2	78.2	55.6	39	81	58	759
IW-4	309.6	128.0	102.0	72.2	41	80	56	477
IW-5	115.5	17.2	10.0	8.5	15	58	49	562
IW-6	163.5	138.0	109.0	94.4	84	79	68	653
IW-7	306.2	50.2	41.9	36.4	16	83	73	577
IW-8	343.8	118.0	105.0	80.8	34	89	68	754
IW-9	114.0	72.0	54.9	47.7	63	76	66	55
Average	254.3	85.5	72.6	60.6	39	82	67	-
Std. dev	133	45	42	39	23	13	17	-

*Averaged value from at least 3 times measurement.

deviation value of 133 mg/Sm^3 and the others were relatively not as scattered with standard deviation values of 45-39 mg/Sm^3. The ratio of PM-10 to TPM showed an average of 39% with wide scattering; in contrast, the ratio of PM-2.5 and PM-1 to PM-10 showed averages of 82% and 67%, respectively, with relatively narrow range. Since most of the coarser particles in TPM were known to be generated from some non-volatile organics in waste or unburned soot by mechanical formation mechanism, they showed a wide range of generation amounts due to different incineration conditions and different contents of ash in the waste. However, most of PM-10 was composed of PM-2.5 or PM-1, which showed the generation and formation of finer particles from nucleation and growth of vaporized metals. As shown in Fig. 1, the ratios of fine particle to coarse particle were slightly increased with flue gas temperature. It could be also inferred that finer particles were generated and grown with nucleation/coagulation/condensation mechanisms. This is discussed in more detail in the following section of this paper for PSD of PM-10 and enrichment of metals in finer particles.

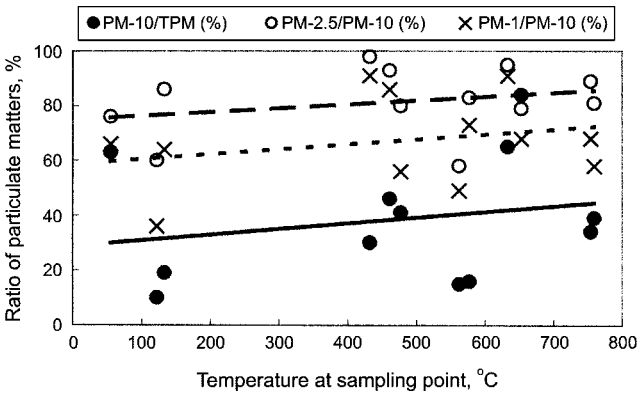

Fig. 1. Ratio of particulate matters with flue gas temperatures.

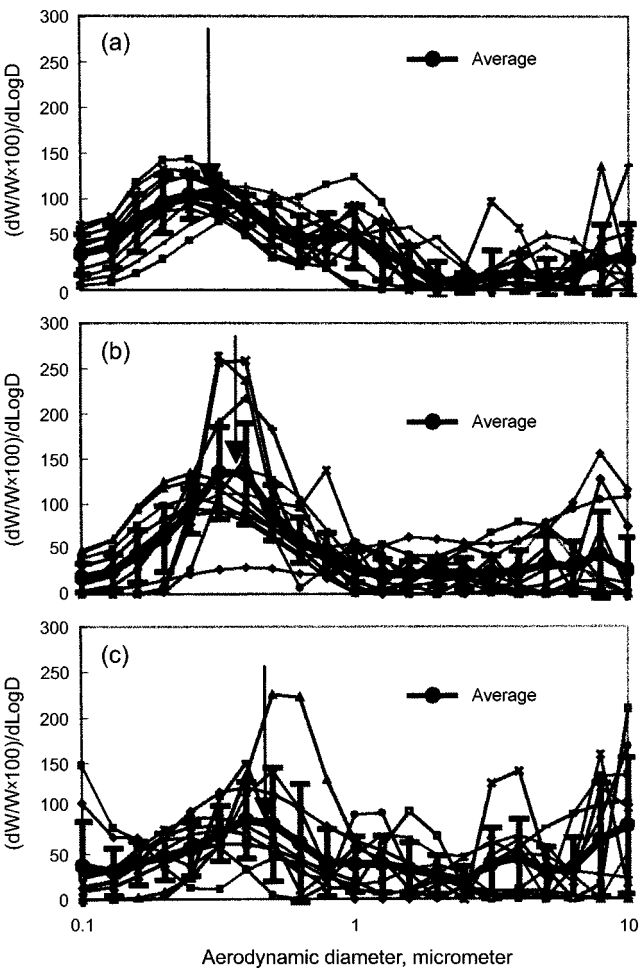

Fig. 2. PSD of PM-10 from small incinerators in different flue gas temperatures. (a) stack temp. >600 °C, (b) 400 °C< stack temp. <600 °C, (c) stack temp. <150 °C.

2. PSDs of PM

The PSDs of PM-10 emitted from the small incinerators are shown in Fig. 2 categorized by flue gas temperature at the stack while temperature of furnace was over 800 °C. As shown in Fig. 2, the distribution of particles was scattered in different incinerators because the emission characteristics were affected by many factors, such as operating conditions, fuel type, efficiency of cyclone, and size of the incinerator. However, the solid line (average distribution) and most of the PSDs showed a peak (mode) between 0.2-0.5 micrometers and the peak shifted to the coarser mode with a decrease in the flue gas temperature (left to right). It suggests that one more mode in the fine particle region exists as other researchers have investigated [US EPA, 2001; Linak et al., 2000; Park and Jeong, 1999; Jang et al., 1996; Kim and Kim, 2002]. The PSD in ambient air is well known to have such bimodal peaks due to their formation and growth by nucleation/condensation/coagulation of vaporized inorganics after emitting from combustion processes such as stationary and mobile sources. Even though they were sampled at APCD outlet, obviously there will be another mode in larger size regions over 5 micrometers as shown in Fig. 2, which started to increase PSD near 10 micrometers.

The fine mode was shifted to bigger size range with temperature decrease as shown in Fig. 2. This could be due to the increased growth by condensation of vaporized metals over existing particles. However, if one considers only coagulation of nucleated particles, higher temperature makes more particle growth due to more collisions of particles, which could be shown by the calculation of the following code (MAEROS2) introduced. Therefore, coagulation could be a dominant mechanism of particle growth in the regime of submicron size while temperature effects must be corrected toward making less particle growth.

Fig. 3 comprises the results of the prediction model of aerosol coagulation, MAEROS [Gelbard and Seinfeld, 1980]. The detailed algorithm can be found in the literatures [Gelbard and Seinfeld, 1980, Linak et al., 1993]. MAEROS was intended as a general tool to supply necessary algorithms to solve the aerosol or particulate system. Originally, the MAEROS code was designed to simulate the dynamics of a spatially homogeneous aerosol. The algorithms can include terms to describe coagulation due to Brownian motion, gravity, and turbulence; particle deposition due to gravitational settling, diffusion, and thermophoresis; particle growth due to condensation of a gas; and time varying sources of particles of different sizes and chemical compositions. The code, MAEROS, also does allow for changes in both pressure and temperature with time, and it is up to the user to configure the algorithms as necessary to apply to a particular environment of interest [Linak et al., 1994]. Fig. 3

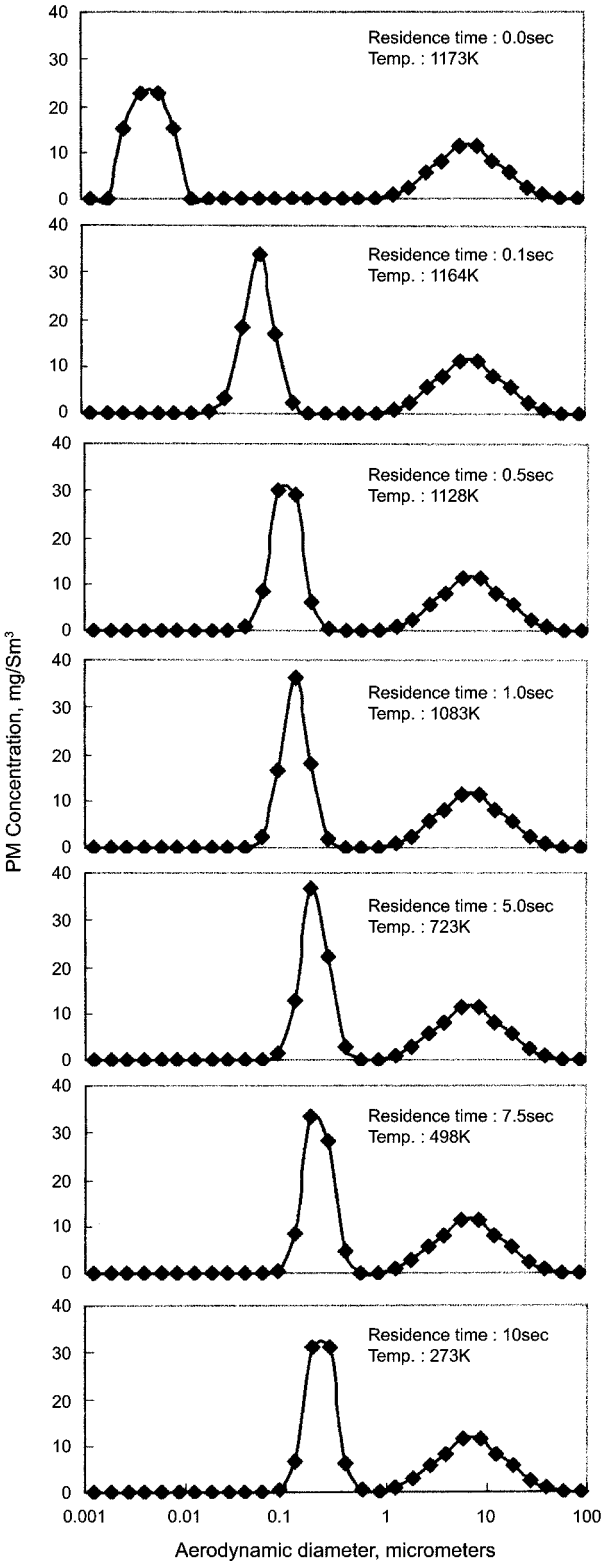

Fig. 3. Predicted evolution of PMs by coagulation in a post-flame combustion environment calculating by MAEROS2 code: Initial concentration in fine mode (0.002-0.01 μm): 75 mg/Sm^3 and coarse mode (1.47-31.6 μm): 58 mg/Sm^3.

illustrates the predicted evolution of an aerosol due to coagulation only.

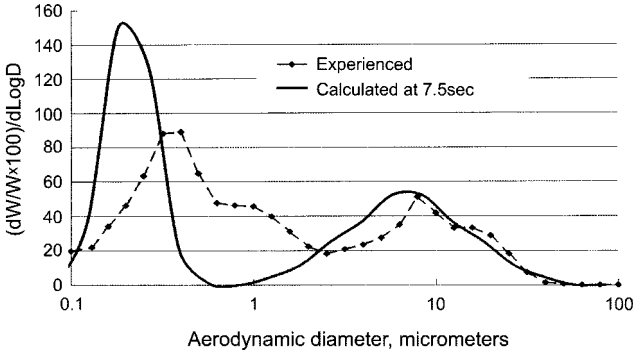

Fig. 4. The experimental PSD in average from small incinerators and model calculated PSD from MAEROS2 code at retention time of 7.5 seconds.

The MAEROS code used, in which the particle diameter size domain (d_p=0.001 to 100 microns), was divided into 30 geometrically equal sections in log-scale. At time zero, an initial mass of 133 mg/Sm^3 was assigned to two modes with lognormal distribution: one was fine mode (d_p=0.002 to 0.01 micron) to simulate the nucleation of a sub-micron fume, and the other was coarse mode (d_p= 1.47 to 31.6 micron) to simulate the mechanically generated particles. The initial mass was estimated from average mass concentration of PM-10 measurement for tested small incinerators, which ranged under 50 microns (cut diameter) as in utilizing a cyclone. Super-micron size PM was considered as mechanically formed and used as the averaged experimental data as input. The PM under 1 micron (PM-1) could be the fine nucleated particles from vaporized metals, and then it would be 75 mg/Sm^3 from the averaged PSD. Temperature was decreased with residence time (1,173 K at initial stage; t=0, to 273 K at time=10 sec) at atmospheric pressure.

The experimental result, in average value of PSD from the small incinerators, and modeling result of retention time at 7.5 seconds are compared in Fig. 4. In the fine mode, the experimental result showed a wider spread than the calculated result, even though the coarse mode was similar in peaks. The differences implied that the other growth mechanism of particulate matters such as condensation, affected the fine particle growth, since the mode could not include condensation behavior.

3. Heavy Metal Emission

The heavy metal concentration in different particle size is shown in Table 3. All the heavy metals excluding chromium and manganese were deposited in the finer particles rather than the larger particles. It can be inferred that the heavy metals, which followed the particle generation mechanisms (i.e., nucleation/condensation/coagulation), were deposited in fine particles. This fact can be explained with reference to PSD in the above section. In the case of chromium and manganese, because of the high melting point and boiling point of chromium species, it was too difficult to vaporize and to follow the particle generation mechanisms.

The characteristics of heavy metal emission from small incinerators were usually affected by hydrogen chloride content in flue gas and flue gas temperature because these factors affected the volatility of various species for each heavy metal [Yang et al., 1994; Jang et al., 2001]. With the assumption of the same heavy metal content in all the wastes for tested incinerators, which is very crude since

Table 3. Heavy metal concentration in different particle sizes (unit: μg/mg)

	Cd		Cr		Cu		Mn		Pb		Zn	
	10-2.5 μm	<2.5 μm	10-2.5 μm	<2.5 μm	10-2.5 μm	<2.5 μm	10-2.5 μm	<2.5 μm	10-2.5 μm	<2.5 μm	10-2.5 μm	<2.5 μm
MW-1	0.5	3.3	78.2	23.7	0.9	2.3	4.6	1.0	2.9	14.8	12.9	108.3
MW-2	2.6	1.3	10.9	1.4	7.0	16.0	2.2	0.0	7.5	11.1	12.9	17.9
MW-3	0.1	2.6	16.8	14.5	0.1	0.1	1.5	1.5	0.7	0.5	1.7	2.9
IW-1	10.0	16.0	6.0	2.0	2.5	6.8	ND	ND	46.0	71.5	35.7	37.2
IW-2	0.2	0.7	3.0	ND	0.8	2.2	ND	ND	10.2	23.0	39.0	117.9
IW-3	1.0	3.7	47.7	6.6	3.6	13.3	0.6	0.3	23.4	115.4	9.9	41.3
IW-4	0.1	1.1	0.04	0.7	0.1	0.4	0.9	0.04	1.0	7.0	0.8	2.5
IW-5	0.05	0.2	0.2	ND	ND	0.1	ND	0.1	ND	ND	12.8	116.1
IW-6	0.1	0.3	21.6	6.3	0.8	2.4	0.2	0.7	1.1	4.0	2.5	3.3
IW-7	0.3	1.2	0.8	ND	0.1	4.1	ND	0.1	2.5	29.5	19.4	115.7
IW-8	37.6	46.6	28.7	2.5	1.9	2.1	2.8	0.1	115.5	187.6	34.7	42.4
IW-9	0.6	3.0	35.5	7.5	0.6	3.3	3.2	1.1	4.5	58.6	2.9	15.9

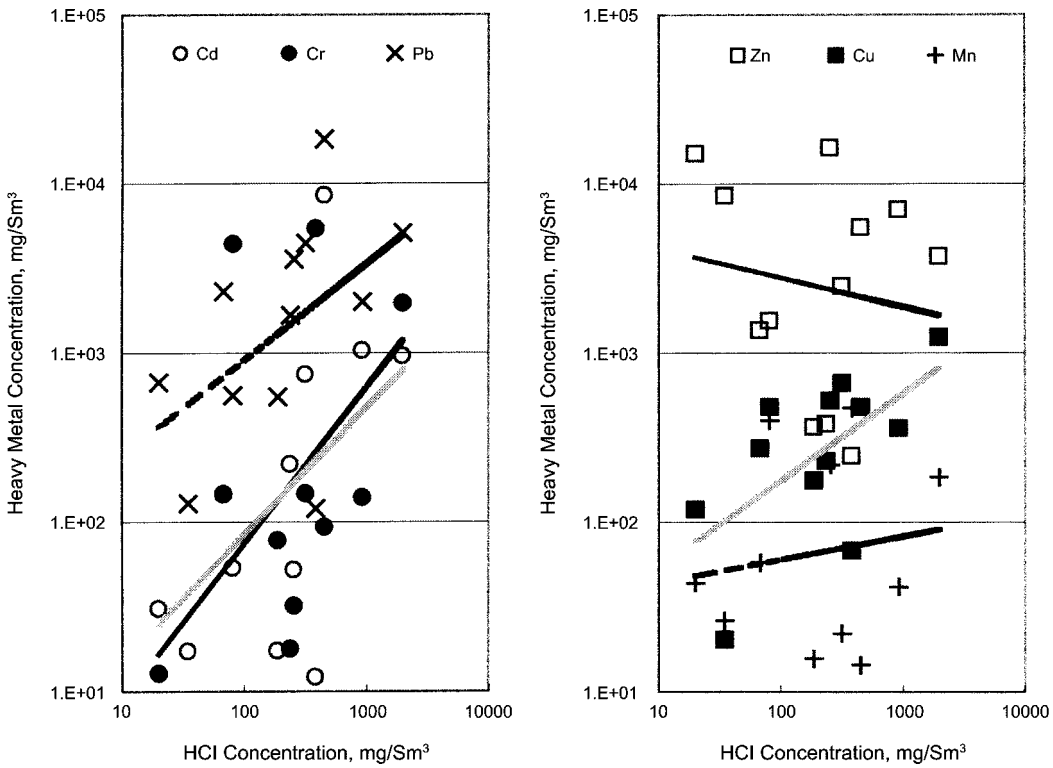

Fig. 5. Emission concentration of heavy metals with respect to HCl concentrations.

tests were carried out at real incinerators, most of heavy metals emission was increased with emitted hydrogen chloride concentration except zinc as shown in Fig. 5. Additionally, in the case of the effect of temperature, as shown in Fig. 6, the emission of manganese, chromium, and copper were not significantly affected by temperature while other heavy metals were increased with temperature. From Fig. 5 and Fig. 6, one can see some effects of temperature and chlorine content. Emission of investigated heavy metals except zinc was affected by hydrogen chloride concentrations, since the chloride compounds of these metals have less vapor pressure than their oxides or elements. Emission of some metals such as manganese, chromium, and copper was not dependent on flue gas temperature.

CONCLUSIONS

The PSD and the heavy metal concentration of flue gas from small incinerators were investigated for characterization of particle growth mechanisms and heavy metal emission with other emission effective factors. Several useful conclusions in the course of characterization of PMs and heavy metals emission could be made. In spite of the fact that the PSD of PM-10 from small incinerators were scattered, the PSD showed a bimodal distribution with a clear fine particle mode under 1 micron, which has been known as metal vapor nucleation, agglomeration/coagulation and condensation as other researchers investigated. Additionally, the particle growth mecha-

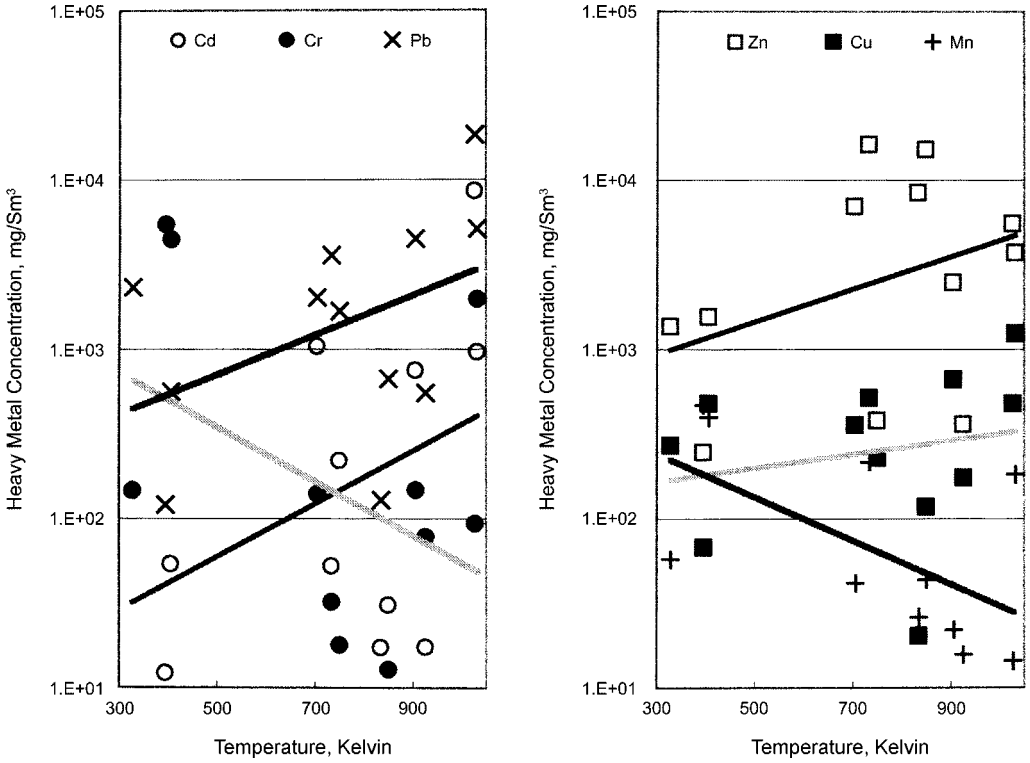

Fig. 6. Emission concentration of heavy metals with flue gas temperatures.

nism, especially coagulation in fine mode, was certified and the PM increases to under 1 micrometer. The portion of fine mode shifted to coarser section with a decrease in flue gas temperature, and retention time increased while the coarse mode was not moved. It could also be explained by particle growth mechanisms. Enrichment of more volatile metals in fine submicron particles could be investigated with supporting such mechanisms. The effects of flue gas temperature and hydrogen chloride concentration in flue gas on the heavy metals emission varied with each heavy metal because they were formed in their different species. Emission of tested heavy metals except zinc was affected by hydrogen chloride concentrations. Some metals such as manganese, chromium, and copper were not dependent on flue gas temperature.

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