

Analysis of heavy metal emission data from municipal waste combustion

Floyd Hasselriis^{a,*}, Anthony Licata^b

^a*Hasselriis Associates, 52 Seasongood Road, Forest Hills, NY 11375-6033, USA*

^b*Licata Energy, 345 Concord Road, Yonkers, NY 10710, USA*

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Abstract

Heavy metals contained in municipal solid waste (MSW), after combustion in modern waste-to-energy facilities, are collected in bottom and fly ash, only a small quantity being discharged from the stack as particulate or vapor. These metals are found to be broadly distributed throughout the constituents, limiting the potential for reducing them by targeting specific components. The many factors which determine metals partitioning to bottom ash, boiler hopper and emission control flyash, and stack emissions, shows that the complex relationship between feed composition and emissions makes it difficult if not impossible to assign cause and effect on their quantities and concentrations in these discharges. Data showing the relationship between particulate matter, emission controls, and emission factors for the heavy metals is examined. A finding that substantial spiking of lead and cadmium in the feed resulted in only marginal changes in stack emissions indicates that efforts to remove these metals from the waste would not produce a significant change in stack emissions. The range of trace metal emissions from a single waste-to-energy (WTE) facility over a period of three to four years is compared with the range reported from individual tests of about twenty facilities also having acid gas controls and fabric filters, indicating that the waste composition and the combustion and emission control technology employed all contribute to the variability of metals and particulate emissions. The relationship between annual averages and probable maximum values which may be anticipated from periodic testing is examined. Special attention is given to mercury, its various species, chemical reactions, and the effectiveness of various carbon-based reagents used for emission control. Emissions from WTE facilities are compared with those from oil and coal-fired utility boilers on a mass per kWh generated.

Keywords: Emissions; Toxic metals; Dioxins; TCDD; Sorbalit; Combustion; Municipal solid waste; Particulate matter; Partitioning of metals; Solubility; Carbon injection; Baghouses; Dry injection; Spray-dry scrubbers

* Corresponding author.

1. Introduction

Municipal solid waste (MSW) contains manufactured and natural materials, including paper, plastics, textiles, food wastes, yard wastes, and other organic materials, as well as inorganic materials such as glass, metals dirt and miscellaneous other components. Almost all of these components contain some quantity of the heavy metals which are categorized as toxic at certain concentrations: lead, cadmium, chromium, mercury, and nickel. From the standpoint of the environmental and health effects resulting from combustion of wastes, it is important to ascertain the quantities, concentrations and chemical forms of the pollutants that are emitted from the stack, both to estimate long-term averages and to anticipate maximum levels of emissions which may occur during compliance tests. Knowledge of the relationship between waste composition and stack emissions may provide useful insight into benefits of source reduction and pre-combustion recycling and removal of components which may make significant contributions to stack emissions.

The analysis which follows reveals the sources of the toxic metals in the waste, and the fractions carried by the combustion gases, removed by the emission control system, and emitted from the stack, and investigates their variability.

2. Metals in the components of municipal solid waste

The elemental metal content of MSW components as well as in the facility discharges has been identified and quantified by landmark tests performed at the Burnaby waste-to-energy (WTE) facility in British Columbia [1]. The median percent of each of the 31 components sampled in the waste is shown in Table 1. Each component was analyzed for 17 metals. By multiplying the concentration of each metal in each component by the weight fraction of the component in the MSW, the contributions of each component to the total metals content can be determined, as shown in Table 1 for cadmium, chromium, mercury and lead.

Examination of Table 1 shows that colored newsprint, residual mixed paper, plastic film, plastic housewares, lawn waste, food containers, and ni-cad batteries were the main sources of cadmium. Chromium was found in the same components, but also in colored newsprint and mixed paper, plastic film, lawn waste, wood, textiles, footwear, and fines (dirt). Mercury was found in paper fractions (perhaps due to fungicides and ink colors), lawn waste, fiberglass and fines, and was derived only to a small extent from alkaline batteries. If the mercury content of these batteries were subtracted, the total mercury content would be reduced by only about 5%. Lead sources were primarily mixed paper, plastic film and housewares, yard waste, wood, textiles, dirt and rocks, small appliances and fines. The lead associated with wood may be natural or from contaminants such as paint. The lead in yard waste may be from atmospheric deposition and uptake from soil. A recent report shows a correlation between the decline in atmospheric lead and the lead content of tobacco grown in Canadian soil, resulting from elimination of leaded gasoline [2]. Many components contributing high levels of lead also exhibit high levels of chromium. Yard waste and certain paper

Table 1
Contribution of components of MSW to metals [1]

			Percent in MSW	Parts per million parts of MSW				
				Cd	Cr	Hg	Pb	
Paper	Fine		2.09	0.002	0.07	0.006	0.09	
	Books		0.24	0.001	0.02	0.000	0.00	
	Magazines	Glued		0.88	0.000	0.15	0.003	0.00
		Not glued		0.93	0.003	0.05	0.003	0.05
	Laminates	Wax/plastic		1.66	0.005	0.05	0.002	0.12
		Foil		0.30	0.000	0.13	0.000	0.28
	Newsprint	Glued		0.29	0.000	0.00	0.000	0.01
		Not - b and w		4.55	0.005	0.17	0.014	0.33
		Color		1.32	0.001	2.84	0.038	0.08
	Browns	Corrugate		9.19	0.009	0.17	0.028	0.35
		Kraft		1.86	0.002	0.09	0.002	0.17
		Box		1.68	0.003	0.09	0.008	0.20
		Mixed paper		13.52	0.230	4.46	0.027	30.96
Plastic	Film	Color	3.13	0.207	3.60	0.013	11.33	
		Flexible	2.51	0.070	2.16	0.005	7.00	
		Rigid	0.3	0.112	0.36	0.001	0.10	
	Food	Pete	0.015	0.001	0.00	0.000	0.01	
		HDPE	0.182	0.005	0.03	0.000	0.11	
		PVC	0.001	0.000	0.00	0.000	0.02	
		DPE	0.001	0.000	0.00	0.000	0.00	
		PP	0.026	0.000	0.01	0.000	0.02	
		PS	0.006	0.000	0.00	0.000	0.00	
		Misc.	0.684	0.542	0.30	0.003	1.08	
	Housewares	Clear	0.064	0.001	0.00	0.000	0.04	
		White	0.262	0.007	1.56	0.001	0.11	
		Blue	0.039	0.113	0.00	0.000	0.03	
		Yellow	0.049	0.001	0.63	0.000	1.21	
		Oher	0.663	0.670	2.38	0.002	4.29	
	Toys, etc.		0.257	0.195	0.59	0.000	0.00	
	Video tape		0.001	0.022	0.00	0.000	0.01	
Organics	Yard	Lawn	10.87	0.652	10.98	0.152	16.74	
		Branches	2.46	0.027	0.59	0.010	1.53	
	Food	Organic	6.76	0.066	0.75	0.010	2.39	
	Wood	Finished	3.29	0.036	3.72	0.007	18.52	
		Unfinished	6.06	0.002	3.51	0.024	19.63	
	Textiles		4.4	0.123	19.36	0.048	5.63	
	Footware		0.65	0.077	11.90	0.001	0.87	
Metals	Ferrous	Beer cans	0.015	0.009	0.05	0.005	0.03	
		Soft drinks	0.012	0.007	0.04	0.004	0.03	
		Food	1.26	0.543	3.64	0.071	4.33	
		Band	0.06	0.009	0.30	0.000	0.36	
	Non-ferrous	Beer	0.058	0.002	0.55	0.000	0.04	
		Soft drink	0.182	0.011	0.16	0.001	0.06	
		Food	0.016	0.000	0.03	0.000	0.02	
		Manufactured	0.40	0.022	5.42	0.001	0.38	
		Foil	0.326	0.166	0.44	0.003	0.00	
		Other	0.001	0.000	0.00	0.000	0.00	

Table 1
Continued

			Percent in MSW	Parts per million parts of MSW			
				Cd	Cr	Hg	Pb
Glass	Combined	Clear	1.52	0.073	0.43	0.003	1.67
		Green	0.12	0.000	1.13	0.000	0.02
		Brown	0.13	0.002	0.06	0.001	0.13
		Other	0.02	0.001	0.02	0.000	0.02
Inorganic light construction		Dirt, rock	0.60	0.120	1.12	0.002	9.27
		Drywall	0.09	0.002	0.01	0.000	0.03
		Fiberglass	0	0.050	14.10	1.100	40.80
		Other	0.87	0.400	34.00	0.100	30.10
Small appliances		Plastic	0.15	0.005	0.38	0.000	0.99
Household batteries		Carbon	0.011	0.003	0.00	0.002	0.00
		Ni-cad	0.007	8.400	0.00	0.000	0.01
		Alkaline	0.012	0.233	0.01	0.029	0.02
Fines			7.6	0.334	8.74	0.106	19.68
Total percent			93.24				
Total parts per million				13.5	93.5	0.73	163.40

fractions, which may be candidates for making compost and recycled paper, contain the highest concentrations of the metals. Elimination of the major sources, batteries, mixed and recycled paper, and yard waste still leaves a significant fraction of the target metals which cannot be removed prior to combustion.

3. Combustion

Combustion converts organic compounds to carbon dioxide and water, hydrogen chloride, sulfur dioxide, nitrogen oxides, carbon dioxide, carbon monoxide, and trace organics. The inorganic matter contained in MSW leaves the system as bottom ash, flyash, vapors or fumes, emerging in chemical and physical forms which are substantially different from those in the waste, as a result of the high temperatures generated, the combustion and emission control technology employed, and chemical reactions due to the presence of oxygen, chlorine, fluorine and sulfur.

3.1 Partitioning of metals during combustion

The partitioning of heavy metals to the bottom ash and flyash during combustion of MSW is determined by many factors, including temperatures in various combustion zones, combustion air distribution, and the physical and chemical form of the waste components containing the metals. The quantity of particulate matter which is carried by the products of combustion rising from the bed of burning solid matter depends upon the velocity of the gases leaving the combustion zone. This velocity

depends upon the shape and configuration of the chambers and the amount and distribution of combustion air. Two-chamber starved-air combustors achieve relatively low carry-over of particulate matter to the combustion products. The carry-over in excess-air stoker-fired combustors depends upon how the underfire and overfire air is distributed. This has been demonstrated by diagnostic tests of the Quebec WTE facility [3].

The heavy metals can be classified according to their degree of volatility. Mercury and arsenic are highly volatile. The presence of chlorine has been found, in laboratory tests, to reduce the volatilization temperatures of nickel, silver, thallium, and lead [4].

As the combustion products leave the furnace and pass through the heat recovery boiler, their temperature is reduced, causing volatile metals such as lead and cadmium, to condense or otherwise attach to flyash. Most of the flyash passes through the boiler to be collected by the air pollution control (APC) system, but substantial amounts stick to boiler tubes. It is periodically removed by soot-blowing or rapping, and deposited in hoppers, later to be collected with the flyash or bottom ash. Deposits formed in the furnace may fall directly into the bottom ash [5].

In addition to furnace temperature, the presence of hydrogen chloride (HCl) affects the amount of lead which is vaporized. Pilot tests showed that increasing HCl from 400 ppm to 1000 ppm increased the quantity of lead vaporized from 2 units to 10 units at 1000 °C, and from 1 unit to four units at 900 °C. The partial pressure of lead oxide (PbO) is substantially lower than that of lead chloride (PbCl₂), which is also influenced by the presence of sulfur [6, 7].

The physical and chemical forms of the waste and of the discharges are important, since they affect toxicity and health effects as well as solubility in water.

Efforts to quantify trace metals partitioning require elaborate testing. It is not possible to obtain simultaneous samples of the waste and the discharges, hence mass balances cannot be closed exactly. It is difficult to quantify and sample the boiler hopper deposits which consist of the particulate matter which sticks to the boiler tubes and is periodically dislodged.

Extensive tests were performed at the Burnaby facility, a water-wall excess-air WTE facility burning MSW, employing humidification and dry lime-injection followed by a fabric filter [1]. The composition of the MSW, boiler outlet flyash and stack emissions are given in Table 2. The metals measured in the boiler exit gases represent only about 6% of the metals measured in the sampled components of the waste, 0.24% of which escaped through the emission controls to the stack. This data indicates that the stack emissions were only about 1/7000 of the metals in the waste.

Uncontrolled particulate leaving the boiler varies with the combustion technology. Particulate emission factors range from about 2 lb/t of MSW for controlled-air (starved-air) combustors to over 30 lb/ton for waterwall combustors and 70 lb/t for RDF combustors, most of which is removed by the APC in any case [8]. Greater amounts of particulate carryover serve to dilute the metal concentration in the flyash [3].

The bottom ash and flyash collected by the various boiler hoppers and the baghouse was weighed and analyzed to construct a mass balance. The analysis of the test data revealed the tendency of the volatile metals, as oxides, chlorides and sulfates

Table 2
Partitioning of metals – tests of Burnaby WTE facility [1]

	Metals in waste (lb/Mt)	Boiler emissions (lb/Mt)	Stack emissions (lb/Mt)	APC control effy. (%)	Range of AP-42 (lb/Mt)
Mercury	1500	3630	1934	46.72	113–3460
Boron	222 000	7496	1370	81.72	
Zinc	3 746 000	249 860	725	99.71	90–725
Lead	326 000	21 681	363	98.33	8–363
Nickel	33 000	1612	105	93.49	2–258
Chromium	185 000	2821	97	96.56	1–210
Tin	98 000	1120	31	97.23	
Cadmium	27 000	5723	18	99.69	3–145
Arsenic	15 800	1048	11	98.95	
Selenium	9600	81	10	87.59	1–10
Vanadium	nm	40	2	95.00	
Copper	284 000	14 508	54	99.63	9–153
Total	4 947 000	305 990	745	99.76	

to collect on boiler surfaces. Similar findings were noted in tests of the RESCO facility in Saugus, MA, which showed the effect of boiler tube rapping cycles on the leachability of metals from the bottom ash, resulting from the highly soluble lead and cadmium chlorides which fall from the boiler tube surfaces [5].

Special tests during which lead-acid batteries, representing about 50 times normal MSW lead content, and cadmium at up to eight times normal levels of cadmium, were fed to the furnace did not show a significant change in stack emissions of these metals. The cadmium was largely collected in hopper and air pollution control (APC) fly ash, and most of the lead in the bottom ash [9]. A conclusion which must be reached from these tests, as well as from the discussion above of the many factors involved, is that there is no direct relationship of cause and effect between the metals content of the waste components and the emissions leaving the stack. There is no reason to believe that a 50% reduction in a specific metal in the waste will reduce its stack emissions by 50%: other factors, such as operating conditions, will have many times as much impact.

The last column in Table 2 shows the range of emissions of WTE facilities having lime reagent-based acid gas controls with fabric filters, listed in the US EPA's AP-42 [10]. The Burnaby data falls in the middle of this range.

4. Emission control systems

Air pollution control systems devices (APCs) collect the particulate matter (PM) which passes through the boiler with the gaseous products. Modern WTE facilities

employ spray-dry or dry lime injection systems to remove the acid gases produced by the sulfur, chlorine and fluorine in the MSW.

4.1. Particle size distribution of particulate matter

The effectiveness of emission controls in removing PM_{10} , particulate which is less than $10\ \mu\text{m}$ in aerodynamic diameter, is of special concern since it can penetrate into the lungs, and since the heavy metals concentrate more on smaller particles due to the greater surface area per unit weight.

Combustion temperatures affect the quantity of metal fumes, less than $1\ \mu\text{m}$ in size, which are produced. Tests of a two-chamber, starved-air refractory combustor burning medical waste showed that increasing the primary furnace temperature from $1350\ ^\circ\text{F}$ to $2150\ ^\circ\text{F}$ increased the percentage of particles in boiler exit flue gases which were less than $1\ \mu\text{m}$ from 15% to 60% [11]. Tests at the WTE facility in Commerce, California, having a water-wall furnace, showed that 6% to 19% of particulate emissions from the fabric filter was less than $1.1\ \mu\text{m}$ [12].

4.2. Reagents used for acid gas control

Reagents injected to absorb and remove acid gases also influence the effectiveness of heavy metals removal along with the particulate matter on which they may be absorbed, adsorbed, chemisorbed, or condensed.

Reducing the flue gas temperatures to $450\ ^\circ\text{F}$, and preferably to $350\ ^\circ\text{F}$ or below enhances chemisorption and adsorption of sulfur dioxide, and organics such as dioxins, furans, chlorobenzenes and chlorophenols, as well as heavy metals, while inhibiting the formation of dioxins which takes place at higher temperatures [13].

4.3. Control efficiencies of various types of APCs

Emission factors, expressed in pounds per million tons of MSW, have been summarized in the US EPA document AP-42 for MSW combustors having various types of air pollution control devices (APCs) [10]. Average emission factors for particulate matter (PM) and the regulated heavy metals, expressed in pounds per million tons of MSW, are listed in Table 3 for electrostatic precipitators (ESPs), spray-dry scrubbers with ESPs (SD/ESP), dry sorbent injection scrubbers with fabric filters (DSI/FF), and spray-dry scrubbers with fabric filters (SD/FF). These emission factors are based on a reference higher heating value (HHV) of 4500 BTU/lb and must be corrected accordingly if other HHV values are involved. The control efficiencies listed were calculated from the average uncontrolled emissions measured at a limited number of facilities, and the average controlled emissions of facilities of each type, for comparison purposes. Since they were not calculated from simultaneous tests of both the APC inlet and outlet gases at the same facility (since both measurements are seldom made) they do not provide a reliable method for predicting emissions of facilities. Nonetheless, Table 3 illustrates the gradation in effectiveness of these control devices: metals control efficiencies calculated from the emission factors range from 99% for ESP's to

Table 3
Particulate matter, metals and acid gas emission factors – AP-42 [10] (Pounds per million tons of MSW)

	No control E.F. ^a	ESP control		SD/ESP control		DSI/FF control		SD/FF control	
		E.F.	Effy (%) ^b	E.F.	Effy (%)	E.F.	Effy (%)	E.F.	Effy (%)
PM	25 100 000	210 000	99.16	70 300	99.72	17 900	99.93	62 000	99.75
As	4370	21.7	99.50	13.7	99.69	10.3	99.76	4.2	99.90
Cd	10 000	646	93.54	75.1	99.25	23.4	99.77	27.1	99.73
Cr	8 970	113	98.74	259	97.11	200	97.77	30	99.67
Hg	4 790	6620	– 38.20!	3260	31.94	2200	54.07	2 200	54.07
Ni	7 850	112	98.57	270	96.56	143	98.18	52	99.34
Pb	213 000	3 000	98.59	915	99.57	297	99.86	261	99.88

^a E.F. = emission factor, pounds per million tons of MSW.

^b Control efficiencies calculated from uncontrolled emissions of various other WTE facilities.

99.9% or higher for facilities equipped with fabric filters, with the notable exception of mercury.

Even when ESPs achieve PM emission levels equal to those of fabric filters, heavy metal emissions are generally substantially higher due to lesser effectiveness in capturing fine particulate, which contains higher concentrations of metal compounds and fumes. Wet scrubbers employing additional devices to condense and remove the escaping vapors and salts have been able to meet stringent PM control requirements, and may prove to be as effective as fabric filters in removing metals.

Inlet/outlet measurements obtained at the same facility provide more valid data for demonstration of the effectiveness of APCs in controlling PM and metals. APC inlet and outlet emission factors obtained from tests at the Burnaby WTE facility, shown in Table 3, permit calculation of APC efficiencies for each specific metal. Subsequent tests show much lower mercury emissions due to injection of activated carbon [1].

Table 4 shows both the uncontrolled and controlled emissions of the Commerce facility in California, a mass-burn WTE facility burning mainly commercial waste, employing a spray-dry scrubber/fabric filter for emission control, based on the average of three tests [12]. The control efficiencies ranged from 91% for mercury, and 97% for beryllium, barium and molybdenum, to 99.99% for lead. Many of the metals emissions were below the detection limit. The absolute emissions for the critical metals were less than about 2 $\mu\text{g}/(\text{Nm}^3)$ (divide pounds per million tons of MSW by 8, or approximately 10 to get $\mu\text{g}/(\text{Nm}^3)$). The mercury emissions of 41 $\mu\text{g}/(\text{Nm}^3)$ were below present and anticipated State and Federal regulatory levels. Table 4 also shows, for comparison, the wide range of emission factors published in AP-42. As can be seen, the Commerce emission factors generally fall in the low side of this range.

4.4. Variability of metal emissions

Individual tests such as those cited above cannot be assumed to represent the variability and range of normal operation and waste characteristics. The emissions

Table 4
Heavy metals collected and emitted – Commerce, California WTE facility [12]

	Boiler emissions ($\mu\text{g}/\text{N m}^3$)	Stack emissions ($\mu\text{g}/\text{N m}^3$)	Control effy (%)	Collected (lb/Mt MSW)	Emitted (lb/Mt MSW)	Range of AP-42 (lb/Mt MSW)
Magnesium	89 933	270	> 99.70	89 663	< 2160	
Barium	4695	117	97.51	4578	936	
Silicon	1860	66	96.45	1794	528	
Calcium	193 000	56	99.97	192 944	448	
Copper	8818	54	99.39	8764	< 432	9–153
Iron	84 167	54	99.94	84 113	< 432	
Mercury	475	41	91.28	434	331	113–3460
Zinc	90 933	38	99.96	90 895	308	90–420
Aluminum	178 000	16	> 99.99	177 984	< 130	
Molybdenum	522	12	> 97.61	510	< 100	
Nickel	4240	6	99.85	4234	50	2–258
Selenium	84	2.7	> 96.76	81	< 22	1–8
Chromium	3620	2.3	99.94	3618	19	1–210
Tin	800	2	> 99.75	798	< 16	
Cadmium	1680	2	99.88	1678	16	3–145
Lead	18 133	2	99.99	18 131	16	8–230
Manganese	3235	1	99.97	3234	8	4–129
Cobalt	111	0.3	99.69	111	3	
Antimony	822	0.3	> 99.96	822	< 2	1–23
Beryllium	7	0.2	> 97.24	7	< 2	0.01–4
Bismuth	31	0.16	> 99.49	31	< 1	
Arsenic	78	0.16	> 99.79	78	< 1	
Vanadium	257	0.09	99.96	257	1	
Total	685 501	745.2	99.89	684 756	5962	

from APC's depend upon the control efficiencies for specific pollutants, which in turn depend upon the inlet concentrations, particle size distribution and chemical forms of the pollutants. Actual emissions are found to vary widely over time, as a result of variations in the metals content in the MSW, and in operating conditions. Metals test data from individual tests often range upward as much as three to five times the average of many tests, as will be seen below. These variations, which result partly from the waste and partly from operational variations, may be great enough to obscure differences between technologies.

4.5. Range of variation in emissions from WTE facilities

The distribution of controlled PM and metals emissions measured annually over a period of three to four years at the Springfield, MA, WTE facility employing dry lime injection fabric filtration is shown in Fig. 1, plotted after sorting the data in

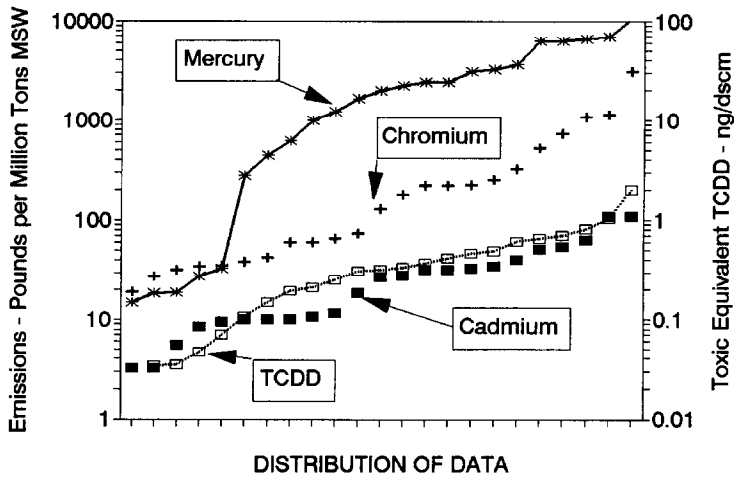


Fig. 1. Distribution of metals emission factors – single WTE facility with dry-injection/baghouse [14].

increasing order. The data, usually measured in micrograms per cubic meter ($\mu\text{g}/\text{m}^3$) can be divided by ten to obtain the approximate pounds per million tons. Both PM and the metals show a range through one to two orders of magnitude, reflecting not only waste composition and equipment variations, but efforts to improve operation. The range of PM is similar to that of mercury and chromium; lead and cadmium show a wider range. In general it can be seen that reductions in PM correspond with equal or increasing reductions in the metals emission factors.

Fig. 2 shows metals emission factors for about twenty WTE facilities with acid gas controls and fabric filters listed in EPA's AP-42, exhibiting the same general trends as the data from the single facility shown in Fig. 1. The twenty facilities experienced roughly the same range of PM as did the single facility, although absolute levels vary.

4.6. Particulate matter as a surrogate for metals

Regulation of PM would indirectly regulate or limit metals emissions if particulate matter can be validly assumed to be a surrogate. PM is measured by passing an iso-kinetically collected flue gas stream through a filter, and weighing the collected solid particulates and condensates accumulated over a period of time. This sample can be analyzed in the laboratory to determine the metals emissions.

From the metals analysis the percentage of these metals in the particulate is calculated. Fig. 3 shows the metals emission factors based on annual compliance test data of the Springfield WTE facility, plotted against the PM data. Individual tests during this period show PM emissions ranging from 0.001 to 0.007 grains per dry standard cubic foot (gr/dscf), corrected to 12% carbon dioxide. Because PM can vary

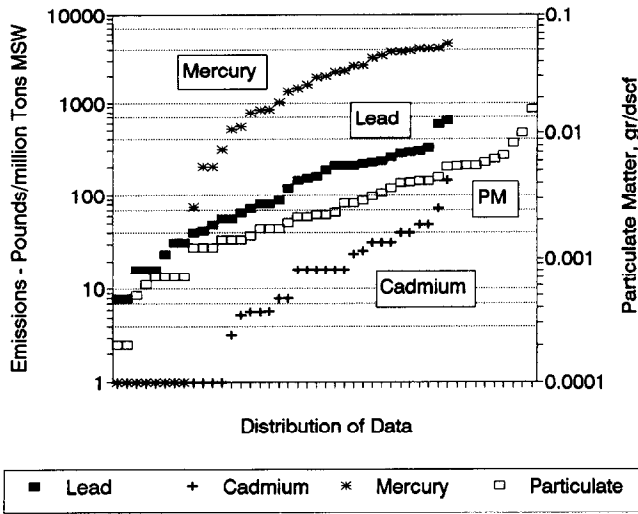


Fig. 2. Distribution of metals emission factors – WTE facilities with scrubber/baghouses [10].

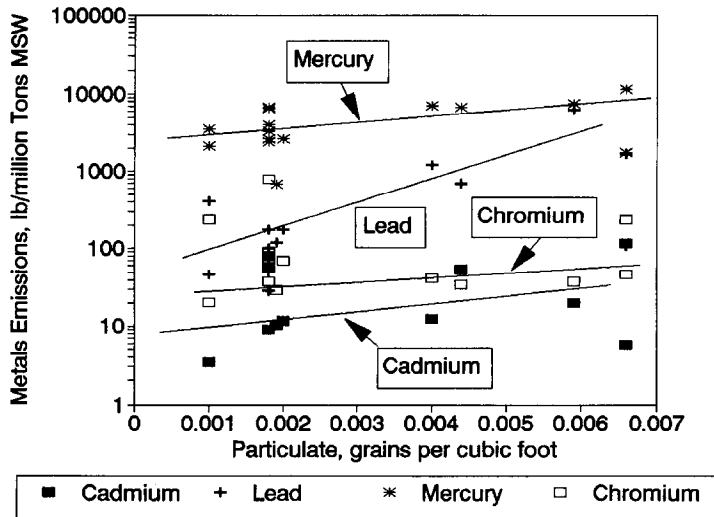


Fig. 3. Metals emission factors versus particulate matter – single WTE facility with dry-injection/baghouse [14].

through this wide range under actual operating conditions, a vendor cannot guarantee performance without sufficient safety margin to allow for this range of variation. In this case, the guarantee could be 0.010 gr/dscf, although the average was closer to 0.004 gr/dscf.

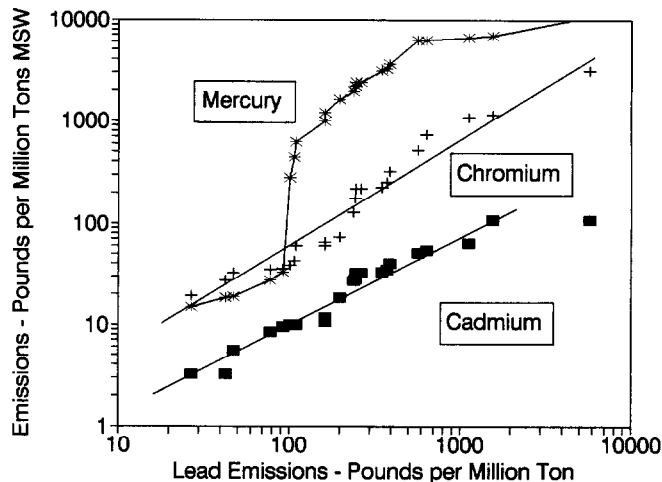


Fig. 4. Metals emission factors versus lead emissions – single WTE facility with dry-injection/baghouse [14].

4.7. Metals emissions versus PM emissions

The emission factors of heavy metals are plotted versus PM in Fig. 3. Cadmium and chromium lie close together, and mercury shows a similar trend. Lead exhibits a sharp decline. These data are consistent with the more effective removal of fine particulate as PM levels are reduced.

4.8. Metals emissions versus lead emissions

Fig. 4 shows emissions of mercury, chromium and cadmium versus lead. It is apparent that cadmium and chromium have a linear relationship to lead. The dip in mercury may be attributed to recycling and changes in operation.

4.9. Metals as a percent of particulate matter

The question as to whether or not PM is a good surrogate for metals can be addressed by examining metals as a percent of PM. The analysis of single facility data is shown in Fig. 5 and that for WTF facilities with scrubber/baghouses in Fig. 6. Mercury was a fairly consistent 7% of particulate, increasing at the 0.001 gr/dscf level to about 15%. Similarly, cadmium was a fairly consistent 0.02% of PM. Lead ranged from about 4% at 0.007 gr/dscf to about 0.5% at 0.001 gr/dscf, a 7-time reduction when PM is reduced 7 times. Chromium showed higher concentrations at low PM emission levels, indicating concentration in the fine particulate. Since these metals show a consistent relationship with PM for a single facility, linear or greater reductions in emissions can be expected as PM emissions are reduced. Therefore, setting an

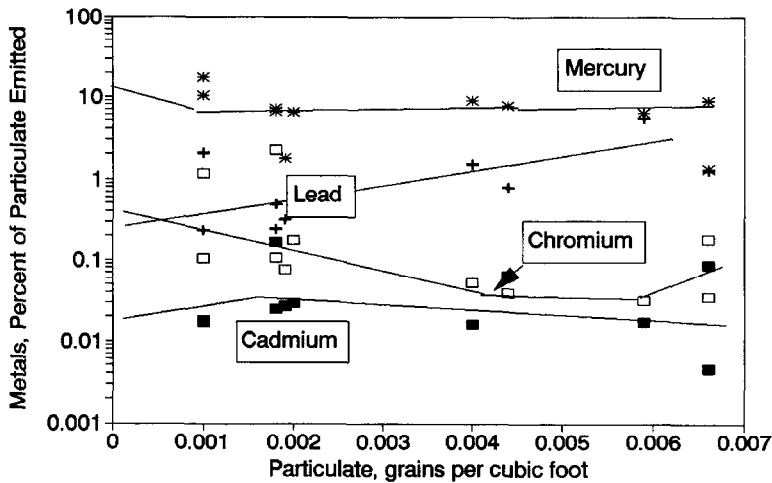


Fig. 5. Metals as a percent of particulate emissions – single WTE facility with dry-injection/baghouse.

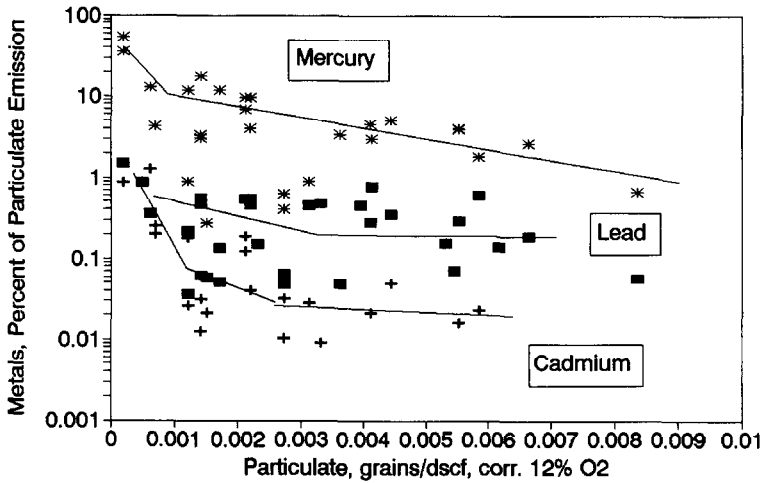


Fig. 6. Metals as a percent of particulate emissions – WTE facilities with scrubber/baghouses.

upper limit for PM emissions provides assurance that metals emissions will be reduced at least in proportion to reductions in PM. It should not be overlooked that actual emissions will average far below the maximum emissions which will occasionally be measured during periodic testing.

4.10. Log-normal characteristic of MSW variation

The distribution of PM and the individual metals in Figs. 1 and 2 shows that the main portion of data falls essentially on a straight line, an indication that the data is

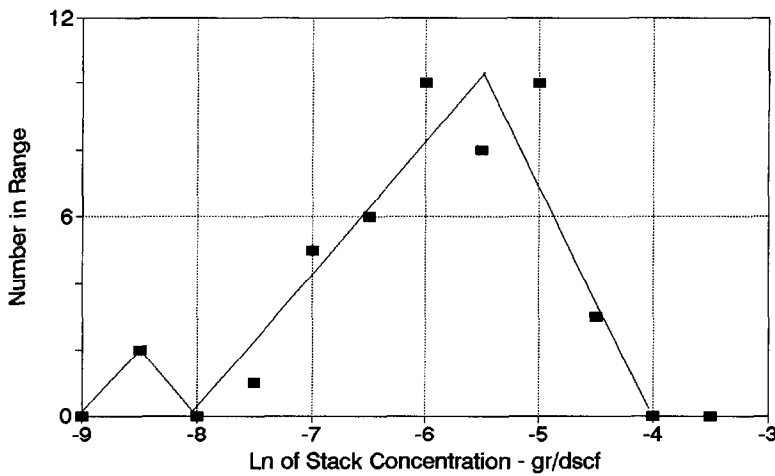


Fig. 7. Log-normal histogram of PM emissions, AP-42 facilities with scrubber/baghouses [10].

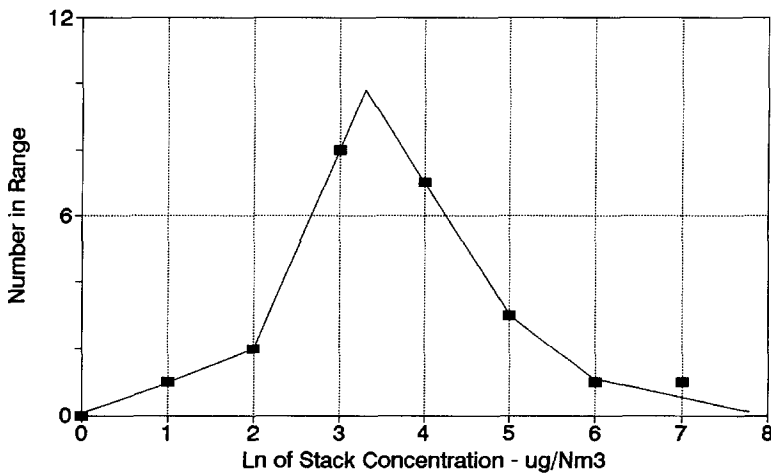


Fig. 8. Log-normal histogram lead emissions, single WTE [14].

log-normally distributed. This indication is confirmed by plotting the histograms shown in Figs. 7 and 8, which exhibit the familiar bell curve of probability. Here the number of test data in each range is plotted against the logarithm of the emission factor. Log-normal distributions are often found for the emissions data of single facilities as well as for groups of facilities having similar but not identical technologies, for some, but not all of the metals.

Fig. 7 shows that PM emissions of the group of AP-42 facilities with scrubber/baghouses exhibit an approximately log-normal distribution. Fig. 8 shows that the histogram of lead emissions of the individual facility exhibits a symmetrical

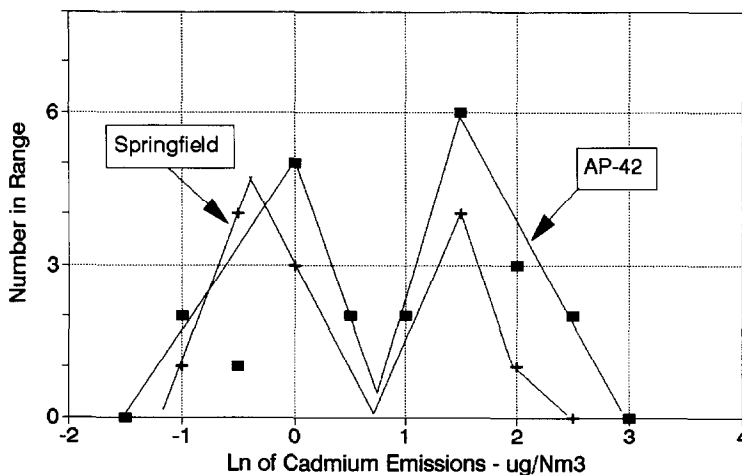


Fig. 9. Log-normal histogram of cadmium emissions, comparing single facility with AP-42 facilities [14, 10].

log-normal distribution. Fig. 9 shows that two separate and remarkably similar distributions of cadmium emissions are found for both the single facility and the AP-42 group, indicating that the characteristics and variations of the waste over time may have a greater effect on emissions than the expected differences in operations and technology.

4.11. Probability graphing

Fig. 10 again shows the data of the single facility obtained over the period of three years, plotted on probability coordinates, which allow extrapolation of the curves to predict the 95% and 99% upper tolerance limits. The trend of particulate matter (TSP) is flat, indicating that 99% of the data would not exceed 0.08 lb/t (about 0.008 gr/dscf), while the probability is that 50% of the data would be less than 0.02 lb/t (about 0.001 gr/dscf). The average of this data is 0.0035 gr/dscf.

The metals follow similar trends except for lead and chromium, which exhibit two slopes, indicating more than one significant population is present. The line for lead extrapolates to 0.06 lb/t at the 99% probability, compared with the 50% point at 10^{-5} lb/t. This wide range may also illustrate the specific impact of the particle size of lead on the performance of emission control devices.

4.12. Estimating annual average and maximum probable emissions

Emissions, measured during individual tests which may be performed at any time during the year, usually average near the middle of the wide range which has been observed during tests of similar WTE facilities.

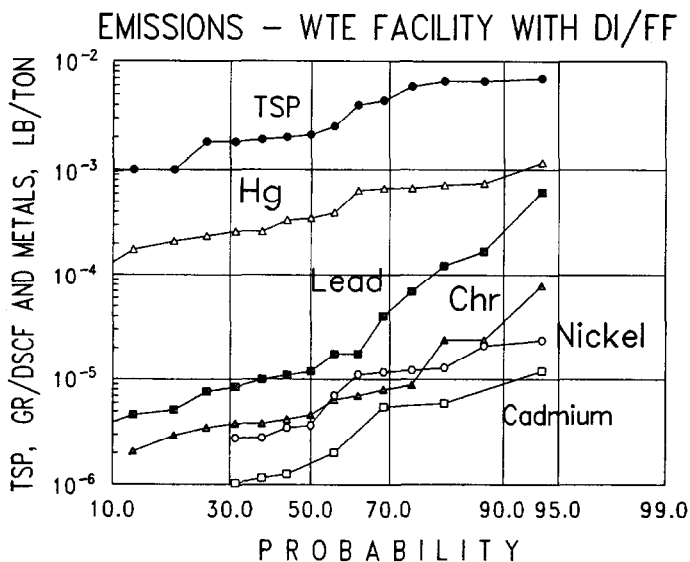


Fig. 10. Log-probability plot of single WTE plant data.

Annual average emissions represent the exposure of the environment to the emissions. As more periodic tests are run, the average of the data becomes more accurately representative of the average impact on the environment. Confidence in the accuracy of this estimate of the average depends upon the standard deviation of the data and the number of test data which have been accumulated. To obtain an estimate of the average, or mean, at a 90% confidence level, the US EPA, in SW-846 (Test Methods for Evaluating Solid Waste) recommends the use of the Upper Confidence Limit, $x + t(s/\sqrt{n})$ where t is the Student's t based on the number of samples, x is the mean, s is the standard deviation, and n is the number of samples or tests [20]. For 20 samples, t is 1.328

As an example, for the lead data used to construct Fig. 6, expressed in pounds per million tons of MSW, the average is 275, the standard deviation is 250, and the UCL for 20 samples is 280. Other data sets may or may not produce similar agreement.

4.13. Maximum probable emissions

There is always a change that one or more tests will measure unusually high emissions. The number below which 90 to 99% of test data fall is a reasonable estimate of maximum probable test results. One way to estimate this number is to add two or three standard deviations to the average (mean) of a set of appropriate data. Test data from WTE facilities show that the standard deviation of various types of pollutants ranges from about 40% to as much as 100% or more of the mean. Hence,

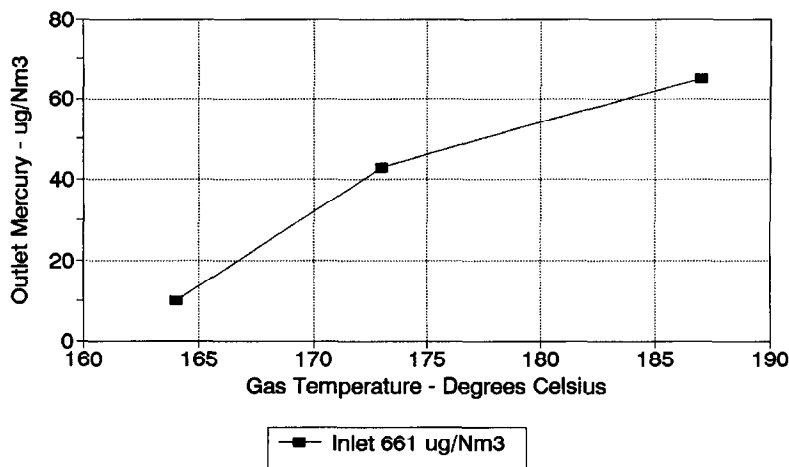


Fig. 11. Mercury emissions – facilities with carbon injection.

the 90 to 99% estimate can easily range from three to five times the mean, depending upon the number of samples [15].

The US EPA draft CETRED document proposes that regulatory emission limits be set on the basis of averages of data from tests of groups of similar facilities [21]. The proposed method for determining the maximum achievable control technology (MACT) for hazardous waste incineration systems uses the Upper Tolerance Limit (UTL) to determine the emission level which will probably not be exceeded in 99 tests out of 100, with a confidence of 95%. This procedure uses a table to determine the constant K , based on the number of samples. The UCL is the mean plus the product of K times the standard deviation. For three samples, K is about 10; for 20 samples, K is about 3.4.

4.14. Mercury emissions from WTE facilities with carbon injection

Fig. 11 shows the range of emissions reported at twelve facilities employing carbon injection. In each case the highest readings were about two times the average, and the lowest were about half the average. This relationship is characteristic of log-normal distributions. Note that all facilities had emissions averaging less than $50 \mu\text{g}/\text{N m}^3$ of mercury.

5. Environmental impact

The environmental impact of WTE facilities includes methods used to manage the ash residues, and the degree of dispersion of the pollutants in the gases which leave the stack.

5.1. Solubility of metals in flyash

The solubility of metals in flyash is a matter of importance for the management of flyash and mixed flyash/bottom ash. The metals in the combustion products may be in the form of oxides, chlorides, sulfates and carbonates. Of these, mainly the chlorides are highly soluble. The presence of chlorine and hydrochloric acid causes a portion of the metals in the flyash to be in a soluble form, causing the flyash to fail the TCLP test for soluble metals.

In spite of the relatively large total metal content of lead, flyash collected from an ESP without acid gas controls was found to release only 4% lead under the conditions of the TCLP leaching test. By comparison, 32% of the cadmium was soluble under these conditions. These two metals are the most likely to cause flyash to fail the TCLP test [5].

Systems employing acid gas controls produce a collected flyash/reaction product mixture which contains a considerable and variable amount of unreacted alkaline product which influences the soluble fraction of lead. Lead is amphoteric, that is, while relatively insoluble when the pH ranges from about 8 to 10, its solubility reaches the EP limit of 5 mg/l at pH less than 5 and greater than 12. Chemical reactions which take place when the flyash is moistened with water can radically change the solubility of lead. For instance, insoluble lead carbonates and phosphates can be formed [22].

5.2. Dispersion of pollutants in stack gases

Dispersion of the stack gases which occurs before the contaminants reach ground levels or sensitive receptors reduces their impact on the environment and human health. Ground level concentrations are predicted by the use of environmental models

Table 5
Comparison of WTE emissions with fossil fuel emissions [25] (lb/1000 MW h)

	Residual oil	Bituminous coal (Pulverized)	Lignite coal (Pulverized)	Waste-to-Energy (Mass burn/refuse derived fuel)
Arsenic (As)	0.22	0.46	0.91	< 0.033
Beryllium (Be)	0.06	0.03	0.06	< 0.017
Cadmium (Cd)	0.18	0.10	0.11	0.063
Chromium (Cr)	0.24	4.56	570	< 0.19
Copper (Cu)	3.19	2.28	3.42	0.43
Mercury (Hg)	0.04	0.23	0.23	0.17
Nickel (Ni)	1436	3.42	3–42	0.84
Lead (Pb)	0.34	0.87	0.11	0.44
Selenium (Se)	NR	0.29	0.29	< 0.022
Vanadium (V)	3.4	4.0	4.0	0.025
Zinc (Zn)	0.47	8.0	8.0	1.23
Particulate	1030	440	440	150

which take into account the stack height, gas exit temperature and velocity, meteorological conditions, and building heights and the characteristics of nearby and remote terrain. The stack gases are diluted by factors which may range from 5000 for incinerators with short stacks and nearby buildings to between 100 000 and over one million for up to 100 m stacks with favorable terrain. Ground-level concentrations of pollutants are generally a few percent or substantially less, than health risk-based acceptable air quality standards [23].

5.3. Comparison of metals emissions from WTE with utility power plants

Table 5 compares the metals emissions, in pounds per 1000 MW-hours of power production for fossil fuels and MSW-burning power plants, based on current regulations regarding emissions of these plants. It is apparent that for the same power output WTE facilities have either similar or much lower emissions of metals [24].

6. Mercury

Emissions of mercury are of concern, especially in areas where high levels are found in the environment. Various countries and individual states have imposed limitations on stack emissions from combustion of MSW. Germany limits emissions to 70 $\mu\text{g}/\text{dscm}$. Florida enacted the first regulation at 70 $\mu\text{g}/\text{dscm}$, or 80% reduction; New Jersey and Minnesota have set limits of 65 $\mu\text{g}/\text{dscm}$, and 80 and 85% reduction, respectively. The US EPA is expected to impose a limit for mercury emitted by new WTE facilities of 100 $\mu\text{g}/\text{dscm}$, or 80% reduction [25].

It has been estimated that natural sources of mercury represent 40 to 65% of the mercury in the global environment, averaging about 5500 t/yr including about 250 t/yr associated with a recent volcano eruption. Man-made, airborne sources of mercury have been estimated to average about 4400 t/yr. About one fifth of this is estimated to come from chlor-alkali facilities, utilities burning coal, oil, natural gas or peat, metal smelters, recycling facilities, geothermal plants, mining drainage and sewage and waste combustion plants [27–29].

An estimate of US sources of mercury emissions from man-made sources, prior to reduction of mercury in batteries, shown in Table 6, includes 33 t/yr, or 4%, coming from MSW combustion [28]. Table 7 shows specific sources of mercury in MSW: in 1985 batteries were reported to account for more than 88% of the mercury in MSW [26]. A 90% reduction to 91 t/yr is projected for the year 2000, primarily as the result of virtual elimination of mercury in household and other batteries.

6.1. Source separation

Mercury will probably escape into the environment from all of our current methods of waste management, landfilling, recycling and WTE, and hence, it would be beneficial to try to mitigate the amount of mercury contained in the waste. Some success has been demonstrated in Hennipen County, Minnesota and Essex County, New Jersey.

Table 6
US Man-made sources of mercury emissions [30]

Source	Mercury emissions (t/yr)
Manufacturing	380
Coal combustion	160
Smelting	70
MSW combustion ^a	33
Fuel oil combustion	1.1
Unidentified	147
Total	790

^a Prior to mandatory mercury reduction in batteries.

Table 7
Sources of mercury in municipal solid waste [29]

	Year 1985		Year 2000	
	(%)	(t/yr)	(%)	(t/yr)
Batteries	88.4	1048	8.8	8
Paints and pigments	4.8	57	11.9	11
Thermometers	2.7	32	18.5	17
Lighting	2.4	28	47.0	43
Thermostats	0.8	9	11.3	10
Dental	0.5	6	2.5	2
All others	0.4	5	—	—
Total	100.0	1185	100.0	91

Battery manufacturers have in the meantime practically eliminated the mercury in alkaline batteries [27].

6.2. Emission control

In addition to reductions by source separation and source reduction efforts, reductions of from 80 to 93% in mercury emissions have been reported at WTE plants employing various forms of carbon reagents. Dioxin reductions from 95 to 99% have been simultaneously reported [16, 17, 25].

6.3. Carbon-based technology

The absorption of mercury and organics such as dioxins into activated carbon and coke is controlled by the properties of both the carbon and the adsorbate, and by the conditions under which they are contacted [16–18]. This phenomenon is generally believed to result from the diffusion of vapor molecules into carbon. These molecules

are retained at the surface in the liquid state because of intermolecular or Van der Waals forces.

As the temperature falls, or as the partial pressure of the vapor above the carbon rises, the average time that a molecule resides on the surface increases. So does the fraction of the available surface covered by the adsorbate. However, the carbon surface is not uniform and consists of sites whose activities vary. More active sites will become occupied first and, as the activity of the remaining available sites decreases, the adsorption energy will change.

The physical structure of activated carbon and coke is not known in detail, but it is believed to contain randomly distributed pores in the carbon, between which lies a complex network of irregular interconnected passages. Pores range in diameter down to a few angstroms, and provide an internal surface area from 300 to 1000 m²/g of carbon. The volume of pores at each diameter is an important variable that directly affects carbon performance.

Laboratory bench scale tests have shown that both increasing the surface area and the addition of sulfur compounds results in higher adsorption rates of elemental Hg. Most of the laboratory work on carbon adsorption has been done on elemental Hg, not with the Hg compounds we normally see in MSW combustor emissions, and without humidification.

Field tests at WTE facilities that inject carbon products with a wide range of surface areas have shown that there is not a significant improvement in the total Hg capture based on the increased carbon surface area. Since high surface area products are more costly, their performance advantages and cost trade-offs have to be demonstrated.

However, the surface area must be available in the proper range of pore sizes. If too much of the area is available in pores smaller than 5 Å, many molecules will be unable to penetrate the pores and that area of the carbon will essentially be unavailable for adsorption. For most pollution-control applications, the surface areas or pores whose diameters range between 5 and 50 Å yield good efficiency rates because the relative pressure of the vapor is usually too low for the larger pores to become filled [16]. At high relative pressures, however, the total pore volume becomes important because the macropores also become active.

The size of the molecules of mercury is approximately 4.5 Å and the dioxin molecules is 10 Å × 3 Å. Both molecules are adsorbed in different parts of the carbon particle. In theory, dioxins are collected in the macropores while the mercury is collected in the micropores. To increase the mercury capture rate the amount of carbon used must be significantly increased, the surface area of the carbon must be increased, or sulfur added.

The carbon/mercury balance has been established through laboratory experiments conducted by Märker Umwelttechnik. They found that under ideal conditions three grams of carbon will absorb one gram of mercury. However, in operating facilities considerably more carbon is required to reduce Hg emissions from 600 µg/dscm to 70 µg/dscm, approximately 300 g of carbon per gram of Hg are used in MWC applications with a baghouse operating at 135°C.

The actual adsorption capacity of carbon is affected by gas temperature, acid content of the flue gas, flue gas moisture, concentration of organics (dioxin), inlet Hg, type of carbon used and surface area, species of Hg and contact time.

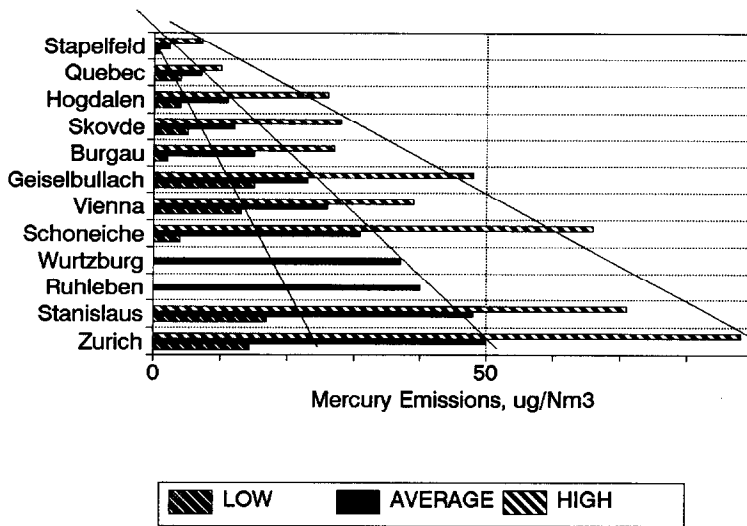


Fig. 12. Mercury removal versus temperature.

The effect of each of these variables has not been quantified. However, through trial and error, the relative effect that flue gas temperature has on Hg adsorption has been demonstrated. A temperature correlation curve is presented in Fig. 12. Another indication of this impact is the carbon adsorption requirement at different temperatures. The following data has been developed by Märker Umwelttechnik at various facilities in Germany using German test methods.

The use of carbon-based technologies was developed in Europe and is now being applied in the US [25]. The most common carbon technology application is the injection of carbon into the flue gas; a second technology is the use of carbon bed filters. Carbon injection technologies are readily applicable to the US WTE industry, as well as to medical waste and hazardous waste combustors. Carbon filter technology requires an extensive air pollution precleaning system before it can be applied to WTE facilities, and requires a relatively high capital investment. To date, this technology has not been demonstrated to be cost effective for US applications.

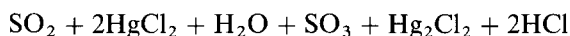
The carbon injection technologies are basically covered by two patents. Niro has licensed their technology to inject 'powdered' carbon to Joy Technologies [19]. The other patented technology is SorbalitTM, a blended product developed by MärkerTM Umwelttechnik of Hamburg, Germany and licensed to Dravo Lime for North American applications [16]. In its simplest form, carbon or SorbalitTM is injected into the flue gas where the carbon component adsorbs mercury and dioxins. An ESP or baghouse located downstream collects the carbon along with other particulate matter. The sorbent component of the technology is produced by mixing lime, either calcium hydroxide or calcium oxide, with surface-activated substances such as activated carbon or lignite coke, and sulfur-based components in a proprietary process.

Sorbalit™ can be produced with carbon contents ranging from 4% to 65% depending on the technical and economic requirements of each project. The process produces an homogeneous powder containing calcium, carbon and sulfur compounds that will not dissociate (demix) when used, either in a slurry or dry form, in the air pollution control systems. A single application of Sorbalit™ will reduce the emissions of SO₂, HCl, dioxins and mercury. Since Sorbalit™ is a blend of lime and carbon, its ignition temperature is significantly higher than that of a carbon product and, therefore, it is a safer material to handle.

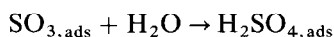
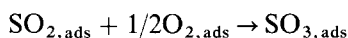
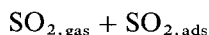
6.4. Theory of sulfur compounds

The addition of sulfur compounds to the process plays a major role in the adsorption of Hg but not in the adsorption of dioxin. Sulfur's role in the adsorption is twofold, first the sulfur compounds maintain the active state of the carbon. Activity is defined as the amount of open pores in carbon. Sulfur's role is to keep these pores open and to allow the Hg to get into the sub-structure pores. The exact process in which sulfur keeps the pores open has not been defined. One theory is that sulfur reacts with water adsorbed or on the surface of the carbon particles to form an acid that penetrates the pores.

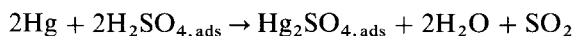
The second role for sulfur is converting elemental mercury (Hg⁰) to a sulfate. Hg⁰ in the vapor phase is more difficult to capture than Hg₂Cl₂ which is the predominate species in MWC emissions. Hg⁰ is about 5–10% of the total Hg emissions from an MWC. Flue gas constituents such as SO₂ can reduce the dissolved HgCl₂ to Hg⁰ which is driven into the gas stream due to its poor solubility.



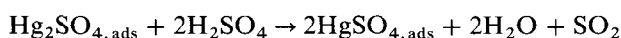
The adsorption capacity of carbon is effected by formation of sulfuric acid on the carbon owing to adsorption of the flue gas constituents SO₂ and H₂O:



Hg⁰ then reacts with the sulfuric acid to form mercurous sulfate (Hg₂SO₄) or in the presence of excess acid mercuric sulfate (HgSO₄):



or



Since the lime component of Sorbalit™ removes the SO₂ from the flue gas some adsorption capacity of the carbon for Hg⁰ is diminished. The sulfur component in Sorbalit™ added during manufacturing replaces the missing SO₂ and enhances the

Table 8
Emission reduction rates [25]

Waste-to-energy plants	Percent reduction	
	Mercury	Dioxins
Marion County, Oregon	87.7	N/M
Würzburg	80–90	99
Schöneiche/Berlin	88	95.6
Schweinfurt	80–93	99

adsorption of Hg⁰. Mercuric chloride does not react with the sulfuric acid but is dissolved in sulfuric acid. No studies concerning the necessary sulfuric acid loading for quantitative precipitation of Hg have been made.

Recent tests have demonstrated the significance that sulfur plays in capturing Hg. The test program conducted at the Marion County, OR MWC showed Sorbalit™ captured more Hg (total and vapor phase) than dry carbon injection [18]. Table 8 shows typical emission reduction rates achieved at WTE plants [25].

In conclusion, activated carbon is effective in reducing mercury and dioxin emissions to the low levels which are now required, or are anticipated in the future.

7. Summary

1. The potentially toxic heavy metals are distributed throughout MSW. Elimination of the major sources, batteries, mixed and recycled paper, and yard waste still leaves a significant fraction which cannot be removed prior to combustion. However, efficient emission controls reduce their discharge to the environment to extremely low levels.

2. The presence of significant amounts of mercury, lead, and cadmium in yard wastes and lawn clippings, as well as in recyclable paper may affect potential for using these materials as feedstock for composting.

3. Efforts to remove specific components in the waste containing high concentrations of metals may not be reflected in significant reductions in stack emissions due to the lack of cause and effect relationships.

4. Data from a single WTE facility supports the assumption that PM is a good surrogate for the heavy metals, since the percentage of the heavy metals in PM was found to be essentially constant through a wide range of corresponding PM emissions. The percentage of lead in PM decreased as PM emissions were reduced.

5. The range of metals emission factors reported from tests of about 20 WTE facilities, listed by the US EPA, having dry or slurry lime injection and baghouses, was isimilar to that measured at a single plant.

6. Metals emissions from a WTE facility over a period of about four years were found to exhibit log-normal distributions for cadmium, chromium and lead; emissions from 20 WTE facilities listed in AP-42 showed a similar range of emissions.

7. The upper confidence limit of data is appropriate for use as a predictor of average annual emissions. For maximum anticipated emissions, the mean plus two or three standard deviations is suggested as a good estimate, since this usually includes about 95% to 99% of the data reported for a large group of facilities.

8. Activated carbon has been found to be capable of removing most of the mercury, dioxins and furans, as well as other trace organic compounds, especially as reduced flue gas temperatures inhibit formation and increase adsorption or chemisorption.

9. The criteria pollutant and metals emissions from WTE facilities, per unit of power generated are shown to be similar to or substantially less than those of residual oil or pulverized lignite or coal-fired utility power plants.

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