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Title: Trace Metal Emission Test Results From A Number Of Industrial And Municipal Point Sources

R. B. Jacko and D. W. Nevendorf

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Trace Metal Particulate Emission Test Results from a Number of Industrial and Municipal Point Sources

Robert B. Jacko and David W. Neuendorf
Purdue University

As part of a Purdue University study of the environmental flow of cadmium and other trace metals, source emission sampling for particulates has been conducted at a number of industrial and municipal plants. The emission samples from each plant were analyzed for Cd, Pb, Zn, Cu, and in some cases Fe and Ni by atomic absorption spectroscopy. Atmospheric mass emission rates and emission factors are reported for total particulates and for each metal. Cadmium emission factors were found to be substantially different from those estimated by Davis in an earlier report. Aerodynamic particle size distributions were measured at each source using an Andersen impactor. A comparison of the total particulate size distributions with the trace metal size distributions revealed that the metals are not evenly distributed among the particle sizes. In every case the trace metals were found predominantly in the respirable particulate fraction even when the bulk of the emissions were non-respirable.

Since 1972 a Purdue University research team has studied various high temperature industrial processes with the objective of characterizing and quantifying the flow of trace metals to and from each process including atmospheric emissions. The plants involved in this cooperative research effort include the East Chicago Municipal Incinerator, rated at 204 metric ton/day of residential and commercial solid waste; a multiple furnace open hearth shop at a Northwest Indiana steel mill producing approximately 8 million metric tons of steel/year; a coker and sinter plant serving a 90,000 metric ton/year vertical retort zinc production facility; and the Purdue University coal fired power plant equipped with 110,000 kg/hr steam boilers.

Dr. Jacko is an Associate Professor of Environmental Engineering in the School of Civil Engineering at Purdue University in West Lafayette, IN 47906. Mr. Neuendorf was a Process Metallurgist with the Cadmium Research Project in the School of Civil Engineering at Purdue University and presently is with Battelle in Columbus, OH.

This paper presents the test results expressed as emission rates and emission factors for total particulate as well as Cd, Pb, Zn, Cu, and in some cases Fe and Ni. Aerodynamic particle size distributions along with observations on the segregation of the trace metals toward respirable particle sizes are also presented.

Description of Facilities Studied

Open Hearth Furnace

The open hearth steelmaking facility has seven 400 ton basic open hearth furnaces, whose exhaust gases are ducted through a common mixing chamber to an electrostatic precipitator (ESP).

The furnaces operate independently (non-synchronously) using a scrap/hot-metal practice common at basic open hearth facilities.[1] The charge proportions range from 10% hot metal/90% scrap to 70% hot metal/30% scrap, depending on the availability of hot metal from the blast furnaces. The types of scrap used range from clean steel scrap to #2 dealer scrap, which includes plated and painted materials. Plated scrap is the prevalent source of the trace metals emitted from the furnaces.[2] The total particulate emissions are a direct function of the amount of oxygen consumed during the heat.[3]

Municipal Incinerator

The incinerator is operated by the City of East Chicago Department of Sanitation, and is the primary refuse processing facility for the cities of East Chicago and Hammond IN (total pop. 154,772).

The plant is equipped with twin furnaces each capable of receiving 204 Mt/day of refuse and are self sustaining after initial gas firing. Atmospheric emissions from each furnace are controlled by a city water-fed spray chamber followed by a three-stage horizontal plate-type scrubbing tower. Throughout the emission tests, the scrubber was operated at its normal liquid to gas ratio of approximately 0.34 liters/m^3.

Residential and commercial refuse contain many sources of trace metal atmospheric emissions.[4] Solder joints in cans and other objects may contain several metals including lead,

Table I. Trace metal particulate emission rate in stack effluent from several sources (kg/hr).

[mean / std. dev.]	Cd	Pb	Zn	Cu	Fe	Ni	Total Particulat
Open hearth furnaces, no control devices (per furnace)	1.28×10^{-1} 3.79×10^{-2}	1.17×10^{1} 3.46	5.19×10^{1} 1.53×10^{1}	4.52×10^{-1} 1.34×10^{-1}	1.20×10^{2} 3.55×10^{1}	1.87×10^{-1} 5.53×10^{-2}	2.77×10^{2} 8.19×10^{1}
Open hearth furnaces, E.S.P. controlled (per furnace)	2.05×10^{-3} 4.39×10^{-4}	1.47×10^{-1} 3.15×10^{-2}	1.00 2.14×10^{-1}	1.10×10^{-2} 2.35×10^{-3}	5.02×10^{-1} 1.07×10^{-1}	2.05×10^{-2} 4.39×10^{-3}	5.58 1.19
Zinc smelter coker, no control devices	1.87 1.15	1.00 6.15×10^{-1}	2.05×10^{1} 1.26×10^{1}	1.57×10^{-2} 9.65×10^{-3}			3.83×10^{1} 2.35×10^{1}
Zinc vertical retorts, no control devices (per retort)	1.38×10^{-2} 4.80×10^{-3}	7.27×10^{-2} 2.53×10^{-2}	1.36 4.73×10^{-1}	4.15×10^{-4} 1.44×10^{-4}			3.03 1.05
Zinc sinter plant, cyclone controlled	1.71×10^{1} 6.13	6.43 2.30	3.88×10^{1} 1.39×10^{1}	1.18×10^{-1} 4.23×10^{-2}			1.91×10^{2} 6.85×10^{1}
Zinc sinter plant, cyclone & E.S.P. controlled	8.57 2.55	5.90 1.75	1.30×10^{1} 3.86	2.74×10^{-2} 8.14×10^{-3}			6.53×10^{1} 1.94×10^{1}
Municipal incinerator, scrubber controlled	5.90×10^{-2} 1.63×10^{-2}	1.20 3.32×10^{-1}	3.40 9.41×10^{-1}	4.50×10^{-2} 1.25×10^{-2}			2.57×10^{1} 7.12
Coal-fired power plant, E.S.P. controlled	1.17×10^{-3}	1.43×10^{-2}	1.07×10^{-1}	6.71×10^{-3}		1.56×10^{-2}	1.65×10^{1}

depending on the application. Plastic objects made of PVC contain cadmium heat stabilizing compounds, and both cadmium and lead are found in paint and ink pigments. Nickel-cadmium batteries can be a significant source of these two metals. Copper brazing is also common in various residential and commercial objects. Thus a wide variety of metals can be expected in the atmospheric particulate effluent from a municipal refuse incinerator.

Zinc Smelter—Vertical Retort

The zinc smelter sintering process, autogenous coking process, and vertical retort reduction process are consecutive stages of the vertical retort zinc refining process[5] used in the plant which was studied. Since cadmium and lead are present as significant impurities in zinc ore, zinc refining is a potential source of atmospheric emissions of cadmium and lead particulate.

The sintering process is intended to remove cadmium from roasted zinc ore, as well as any sulfur that remains after the roasting process. Roasted ore (calcine) is mixed with a controlled amount of powdered coal and sintered. Exhaust gases containing particulate enriched in cadmium and lead, pass through a pair of cyclones, a water spray chamber and an ESP before being discharged to the atmosphere.

The next step, the autogenous coking process, involves mixing the sintered product with other zinc bearing materials, briquetting the mixture and coking the resulting briquets. The object of coking is to produce briquets with as much as possible of their original ZnO content in a mechanically strong coke matrix, free of the volatile organics that would foul the vertical retorts. The coker combustion products are released through an uncontrolled stack and can include copious metallic fume.

Coked briquets are transported within seconds to a vertical retort, where zinc, lead, and cadmium vapors are extracted in a reducing atmosphere. The combustion of natural gas and recycled CO in a chamber surrounding the retort maintains it at 1300°C. Gases containing particulate matter leave the combustion chamber through a recuperator (used for preheating combustion air) and are released to the atmosphere through an uncontrolled stack. This stack serves a battery of nine vertical retorts. A more detailed description of the processes sampled including drawings can be obtained directly from the authors.

Coal-Fired Power Plant

The Purdue power plant is equipped with two coal-fired boilers, each rated at 110,000 kg/hr of steam, one of which was sampled for this study. Each Wick boiler is fired with high sulfur Indiana coal via spreader stokers. Atmospheric particulate emissions are controlled by a Research Cottrell ESP. The resulting process steam is used for space heating and air conditioning.

Sampling Procedures

All particulate samples were taken using the standard E.P.A. sampling train.[6] In each case the train was modified by the use of 5% nitric acid in the first two impingers, in order to ensure capture of any trace metal particulate which might have bypassed the glass fiber filter.[7] The glass fiber filter, probe backwash, and impinger contents were analyzed for trace metals by atomic absorption (AA) spectroscopy after digestion in concentrated HNO_3. The AA unit used was a Perkin-Elmer model equipped with a deuterium background corrector.

Aerodynamic particle size samples were taken with the Andersen high temperature stack head[8] fitted to the nozzle end of the sampling probe. All Andersen samples were removed from the plates and analyzed in the same manner noted above for particulate samples.

Traverse points for particulate sampling were selected according to E.P.A. Method 1.[6] Andersen samples were taken at constant isokinetic sample flow rate at an average velocity point in each stack.

Trace Metal Particulate Emissions

Tables I and II summarize the atmospheric mass emission rates and emission factors, respectively, for the trace metals analyzed. Since each value reported is the average of a number of samples, the standard deviation is also shown to indicate variations attributable to the process or the test procedure. The probable error in E.P.A. method 5 stack testing proce-

Table II. Trace metal particulate atmospheric emission factors from several sources.

[mean / std. dev.]	Cd	Pb	Zn	Cu	Fe	Ni	Total Particulate
Open hearth furnaces, no control devices (kg/Mt steel)	2.89×10^{-3} 8.54×10^{-4}	2.63×10^{-1} 7.78×10^{-2}	1.17 3.46×10^{-1}	1.02×10^{-2} 3.02×10^{-3}	2.70 7.98×10^{-1}	4.23×10^{-3} 1.25×10^{-3}	6.25 1.85
Open hearth furnaces, E.S.P. controlled (kg/Mt steel)	5.50×10^{-5} 1.17×10^{-5}	3.95×10^{-3} 8.42×10^{-4}	2.70×10^{-2} 5.76×10^{-3}	2.95×10^{-4} 6.29×10^{-5}	1.35×10^{-2} 2.88×10^{-3}	5.50×10^{-4} 1.17×10^{-4}	1.50×10^{-1} 3.20×10^{-2}
Zinc smelter coker, no control devices (kg/Mt zinc)	1.17 7.18×10^{-1}	6.25×10^{-1} 3.83×10^{-1}	1.28×10^{1} 7.85	9.82×10^{-3} 6.03×10^{-3}			2.39×10^{1} 1.47×10^{1}
Zinc vertical retorts, no control devices (kg/Mt zinc)	3.25×10^{-2} 1.13×10^{-2}	1.69×10^{-1} 5.86×10^{-2}	3.20 1.11	9.77×10^{-4} 3.39×10^{-4}			7.15 2.48
Zinc sinter plant, cyclone controlled (kg/Mt zinc)	3.16 1.13	2.18 7.82×10^{-1}	4.80 1.72	1.01×10^{-2} 3.62×10^{-3}			2.41×10^{1} 8.64
Zinc sinter plant, cyclone & E.S.P. controlled (kg/Mt zinc)	1.08 3.21×10^{-1}	7.45×10^{-1} 2.21×10^{-1}	1.64 4.87×10^{-1}	3.46×10^{-3} 1.03×10^{-3}			8.25 2.45
Municipal incinerator, scrubber controlled (kg/Mt refuse)	9.0×10^{-3} 2.5×10^{-3}	1.9×10^{-1} 5.3×10^{-2}	5.0×10^{-1} 1.4×10^{-1}	7.0×10^{-3} 1.9×10^{-3}			4.0 1.1
Coal-fired power plant, E.S.P. controlled (kg/Mt steam)	2.38×10^{-5}	2.92×10^{-4}	2.19×10^{-4}	1.37×10^{-4}		3.19×10^{-4}	3.37×10^{-1}

dures is ±10.4%,[9] therefore, standard deviations greater than 10.4% of the mean value are process related. For example, the standard deviation of the uncontrolled open hearth cadmium emission rate in Table I is approximately 30% of the mean and is therefore attributable predominantly to the process and not to the test procedure. Also note in examining Table I and II that the trace metal and total particulate emissions were substantially reduced with effective control equipment.

The open hearth steelmaking process does emit trace metal particulate as expected, but the metals are effectively removed from the exhaust gas by an electrostatic precipitator. Trace metal removal was achieved at a level of 98% by mass (Table I). The average Cd emission factor (2.89×10^{-3} kg/Mt-steel, Table II) for the uncontrolled open hearth furnace is 0.4 times the value (7.5×10^{-3} kg/Mt) estimated by Davis, *et al.*[10] for uncontrolled iron and steel production. The combination of the factor of 0.4 and the fact that many open hearth furnaces have control devices may result in substantially lower environmental impact of Cd emissions than estimated by Davis for steelmaking. The use of basic oxygen furnaces which are equipped with at least as efficient a control device as tested here should further reduce the overall cadmium emission factor for the steel industry.

The vertical retort zinc smelting process releases 1.08 kg-Cd/Mt-Zn at the sintering stage, 1.17 kg-Cd/Mt-Zn at the coking stage and 3.25×10^{-2} kg-Cd/Mt-Zn at the vertical retort reduction stage. These emission factors, which are based on Zn flow through the individual process steps, can be used to calculate an overall Zn smelting emission factor based on production rate of Zn metal. To do so, the emission factor for each stage must be multiplied by the ratio of Zn flow through the stage to total Zn production, in order to correct for losses of Zn in various parts of the process. Finally the modified emission factors are summed to produce an overall emission factor. The result is an emission factor of 1.48 kg-Cd/Mt-Zn produced. This factor is 1.80 times as large as the value estimated by Davis, *et al.*[10] Davis assumed that all phases of Zn smelting are equipped with control devices. If the coker studied here had been controlled by a baghouse or ESP, the two emission factors would be in closer agreement.

The East Chicago Municipal Incinerator produces 9.0×10^{-3} kg-Cd/Mt-refuse, which is 6.0 times the 1.5×10^{-3} estimated by Davis.[10] This difference is magnified in importance by the fact that Davis assumed no control devices on incinerators, whereas the East Chicago unit is controlled by a low energy spray-type scrubber. Thus the release of Cd to the atmosphere by refuse incineration (as opposed to sewage sludge incineration) is greater than that estimated by Davis.

Aerodynamic Particle Size

Aerodynamic particle size distributions for each process are presented in Figure 1. Mass median diameters (diameter at the 50% point on the % less than scale) range from 0.84μm for the open hearth ESP outlet to 5.1μm for the coal-fired power plant ESP outlet.

For all the processes studied, it was observed that a high percentage of the trace metals were emitted as relatively small

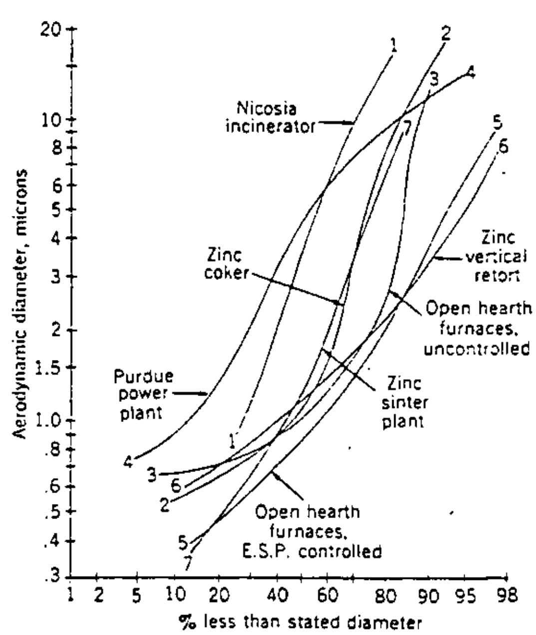

Figure 1. Aerodynamic particle size distributions of industrial stack effluents, log-probability plots.

particles. Figures 2 through 5 illustrate this segregation trend for Cd, Pb, Zn, and Cu in each process. For each metal the percentage of the metal which is found on a given Andersen impaction stage is plotted against the average aerodynamic diameter of particulate on that stage. A similar phenomenon has been observed for a coal fired electric generating plant.[11,12]

Note that this segregation is very pronounced for the relatively volatile metals, Cd and Pb. The effect is still noticeable for Zn which is less volatile, but diminished for Cu, the least volatile of the four metals. This trend is consistent with two proposed mechanisms, each of which requires that the metals pass through the vapor phase. One is the generation of metallic fume. The other is a surface adsorption mechanism whereby the metallic vapor condenses on the surface of a predominantly non-metallic substrate particulate.[13]

Tables III and IV separate the trace metal particulate mass emission rates and emission factors into their respirable and non-respirable fractions. Particles above 2μm aerod[illegible]nic diameter are considered non-respirable (not penetratin[illegible] [illegible]nd being retained by the alveoli of the lungs). Particles smaller than 2μm are assumed to be respirable.[14,15]

Note that in some cases there are contrasts between the proportions of total particulate and trace metals that are *less than* (<) versus *greater than* (>) 2μm. At the municipal incinerator, for example, 1.54 kg of <2μm and 38.5 kg of >2μm particulate are emitted per Mt of refuse incinerated. This means that 96% of the total particulate emitted is classified as non-respirable. On the other hand, the metals are found to a greater extent in the respirable fraction than the non-respirable. Note that 66% of the Cd, 88% of the Pb, 71% of the Zn, and 82% of the Cu are in the respirable fraction.

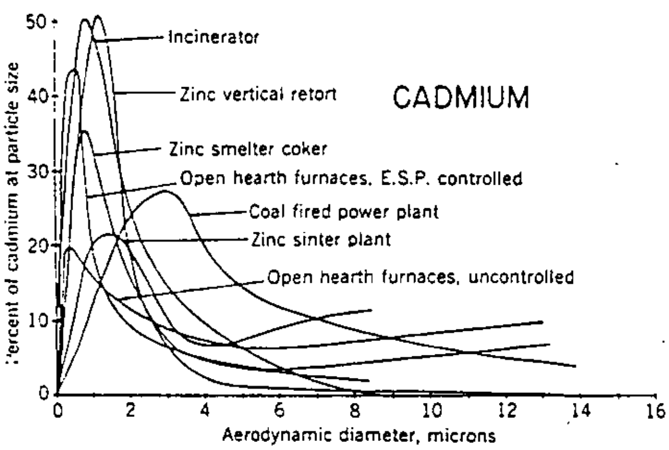

Figure 2. Aerodynamic particle size distribution of cadmium in industrial stack effluent.

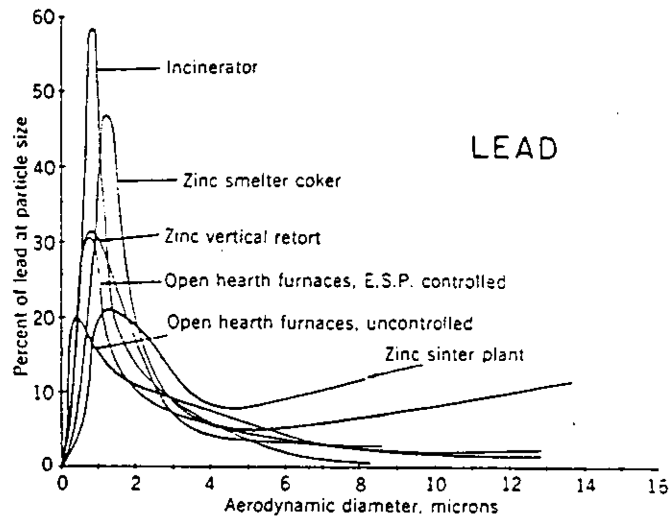

Figure 3. Aerodynamic particle size distribution of lead in industrial stack effluent.

Table III. Emission rates of trace metal particulate in stack effluent greater and less than 2μm aerodynamic diameter (kg/hr).

		Cd	Pb	Zn	Cu	Fe	Ni	Total Particulate
Open hearth furnaces, no control devices	(R)[a]	6.55×10^{-2}	8.89	3.97×10^{1}	3.19×10^{-1}	9.42×10^{1}	5.95×10^{-2}	1.98×10^{2}
(per furnace)	(N)	6.25×10^{-2}	2.81	1.22×10^{1}	1.33×10^{-1}	2.58×10^{1}	1.28×10^{-1}	7.90×10^{1}
Open hearth furnaces, E.S.P. controlled	(R)	1.59×10^{-3}	1.14×10^{-1}	7.15×10^{-1}	8.56×10^{-3}	4.25×10^{-1}	1.30×10^{-2}	4.47
(per furnace)	(N)	4.60×10^{-4}	3.30×10^{-2}	2.85×10^{-1}	2.44×10^{-3}	7.70×10^{-2}	7.50×10^{-3}	1.11
Zinc smelter coker, no control devices	(R)	1.20	6.00×10^{-1}	3.98	6.94×10^{-3}			1.87×10^{1}
	(N)	6.70×10^{-1}	4.00×10^{-1}	1.65×10^{1}	8.76×10^{-3}			1.96×10^{1}
Zinc vertical retorts, no control devices	(R)	1.15×10^{-2}	5.66×10^{-2}	9.67×10^{-1}	1.77×10^{-4}			2.14
(per retort)	(N)	2.30×10^{-3}	1.61×10^{-2}	3.93×10^{-1}	2.38×10^{-4}			$8.90 \times 10^{-}$[illegible]
Zinc sinter plant, cyclone & E.S.P.	(R)	5.14	3.45	6.29	1.09×10^{-2}			2.59×10^{1}
controlled	(N)	3.43	2.45	6.71	1.65×10^{-2}			3.94×10^{1}
Municipal incinerator, scrubber con-	(R)	3.90×10^{-1}	1.05	2.42	3.71×10^{-2}			9.87
trolled	(N)	2.00×10^{-2}	1.5×10^{-1}	9.80×10^{-1}	7.90×10^{-3}			1.58×10^{1}
Coal-fired power plant, E.S.P. con-	(R)	3.72×10^{-4}		2.91×10^{-2}	2.15×10^{-3}		5.76×10^{-3}	2.89
trolled	(N)	7.98×10^{-4}		7.79×10^{-2}	4.56×10^{-3}		9.84×10^{-3}	1.36×10^{1}

[a] R: respirable fraction (<2μ aerodynamic diameter)
N: non-respirable fraction (>2μ aerodynamic diameter)

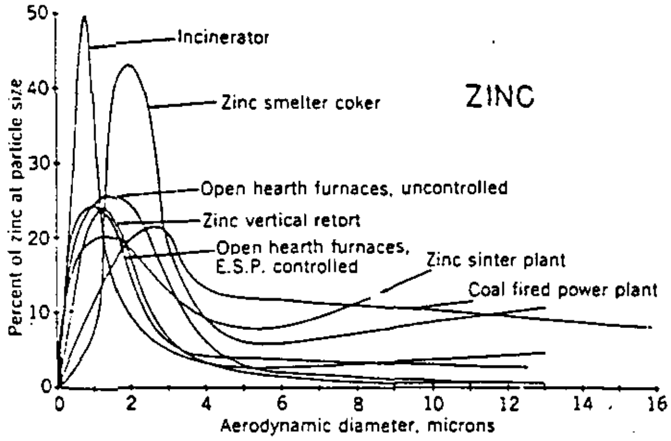

Figure 4. Aerodynamic particle size distribution of zinc in industrial stack effluent.

Conclusions

The open hearth steelmaking process, vertical retort zinc smelting process, municipal refuse incinerator and coal-fired power plant are all sources of measurable trace metal atmospheric emissions. Much of the metal is released in an aerodynamic size range below 2μm diameter.

Specific conclusions follow:

1. The open hearth steelmaking furances controlled by an ESP released 0.15 kg of particulate per Mt of steel produced. Approximately 80% of this total particulate was found to be <2μm aerodynamic diameter along with 78% of the Pb and Cd.
2. The uncontrolled open hearth furnaces had total particulate emissions of 6.3 kg/Mt-steel. About 72% of the total particulate, 51% of the Cd and 76% of the Pb was <2μm aerodynamic diameter.
3. The zinc smelter uncontrolled briquet coker emitted 24 kg of total particulate per Mt of Zn in the briquets. Only 49% of the total particulate emissions was <2μm aerodynamic diameter compared to 64% of the Cd and 60% of the Pb.
4. The vertical zinc retorts (with no control equipment on the firing chamber exhaust) released 7.2 kg-particulate/Mt-Zn produced. 70% of the total particulate, 83% of the Cd and 78% of the Pb were <μm aerodynamic diameter.
5. The sintering machine at the zinc smelter, controlled by cyclones and an ESP, emitted 8.3 kg/Mt-Zn total particulate. Only 40% of this total particulate was <2μm, while 60% of the Cd and 58% of the Pb was found to be <2μm.

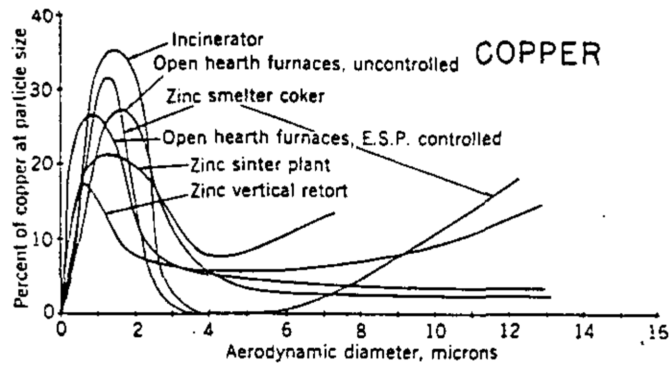

Figure 5. Aerodynamic particle size distribution of copper in industrial stack effluent.

6. Of the 40 kg-particulate/Mt-refuse released by a scrubber controlled municipal incinerator, only 4% was <2μm. In contrast, 66% of the Cd and 88% of the Pb was found to be <2μm aerodynamic diameter.
7. The ESP controlled coal-fired power plant produced 0.34 kg-particulate/Mt-steam, of which 18% was <2μm. However, 32% of the Cd was contained in the respirable fraction.

Thus bulk particulate metal analyses and aerodynamic particle size determinations have been shown to be insufficient

Table IV. Atmospheric emission factors for trace metal particulate greater and less than 2μm aerodynamic diameter

		Cd	Pb	Zn	Cu	Fe	Ni	Total Particulate
Open hearth furnaces, no control devices	(R)[a]	1.48×10^{-3}	2.00×10^{-1}	8.94×10^{-1}	7.20×10^{-3}	2.12	1.35×10^{-3}	4.47
(kg/Mt steel)	(N)	1.41×10^{-3}	6.30×10^{-2}	2.76×10^{-1}	3.00×10^{-3}	5.80×10^{-1}	2.88×10^{-3}	1.78
Open hearth furnaces, E.S.P. controlled	(R)	4.26×10^{-5}	3.07×10^{-3}	1.93×10^{-2}	2.30×10^{-4}	1.14×10^{-2}	3.49×10^{-4}	1.20×10^{-1}
(kg/Mt steel)	(N)	1.24×10^{-5}	8.80×10^{-4}	7.70×10^{-3}	6.50×10^{-5}	2.10×10^{-3}	2.01×10^{-4}	3.00×10^{-2}
Zinc smelter coker, no control devices	(R)	7.49×10^{-1}	3.75×10^{-1}	2.48	4.34×10^{-3}			1.17×10^{1}
(kg/Mt zinc)	(N)	4.21×10^{-1}	2.50×10^{-1}	1.03×10^{1}	5.48×10^{-3}			1.22×10^{1}
Zinc vertical retorts, no control devices	(R)	2.70×10^{-2}	1.31×10^{-1}	2.28	4.16×10^{-4}			5.04
(kg/Mt zinc)	(N)	5.50×10^{-3}	3.80×10^{-2}	9.20×10^{-1}	5.61×10^{-4}			2.11
Zinc sinter plant cyclone & E.S.P. controlled (kg/Mt zinc)	(R)	6.48×10^{-1}	4.35×10^{-1}	7.94×10^{-1}	1.37×10^{-3}			3.28
	(N)	4.32×10^{-1}	3.10×10^{-1}	8.46×10^{-1}	2.09×10^{-3}			4.97
Municipal incinerator, scrubber controlled (kg/Mt zinc)	(R)	5.95×10^{-3}	1.67×10^{-1}	3.56×10^{-1}	5.77×10^{-3}			1.54
	(N)	3.05×10^{-3}	2.30×10^{-2}	1.44×10^{-1}	1.23×10^{-3}			3.85×10^{1}
Coal-fired power plant, E.S.P. controlled (kg/Mt steam)	(R)	7.57×10^{-6}		5.96×10^{-5}	4.40×10^{-5}		1.18×10^{-4}	5.90×10^{-2}
	(N)	1.62×10^{-5}		1.59×10^{-4}	9.30×10^{-5}		2.01×10^{-4}	2.78×10^{-1}

[a] R: respirable fraction (<2μ aerodynamic diameter)
N: non-respirable fraction (>2μ aerodynamic diameter)

data for complete characterization of trace metal particulate emissions. The degree of metal segregation into the respirable particulate fraction should also be determined by chemical analysis of the particle size samples. This information will be useful both for environmental impact assessment and for design of control equipment for removal of <2μm aerodynamic diameter particulate which contain a substantial fraction of the trace metals in stack effluents.

Acknowledgments

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