United States
Environmental Protection
Agency

Environmental Sciences
Research Laboratory
Research Triangle Park NC 27711

Research and Development

EPA-600/S3-84-047 May 1984

EPA

Project Summary

Chemical and Physical Characterization of Municipal Sludge Incinerator Emissions

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Particulate emissions from a group of municipal sludge incinerators, three with multiple-hearth furnaces and one with a fluidized-bed furnace, were characterized in a study covering a period from October 1, 1979 to June 30, 1981. Objectives of the investigation were (1) to obtain specific elemental emission concentrations, and (2) to provide source inventories and source signatures, especially in terms of particle size, that would assist in developing and evaluating source apportionment models. Three of the plants investigated in this study operated at or near autogenous burning conditions. Chemical element composition was determined for total and sized emission samples by x-ray fluorescence analysis. During the study, considerable enrichment of several elements (S, V, Cu, Zn, Cd, Sn and Pb) in the emissions, compared to their content in the sludge feed, was observed. The largest average enrichment ratios were found with cadmium (31), zinc (14), lead (9), and sulfur (8).

This Project Summary was developed by EPA's Environmental Sciences Research Laboratory, Research Triangle Park, NC, to announce key findings of the research project that is fully documented in a separate report of the same title (see Project Report ordering information at back).

Introduction

With the production of municipal wastewater sludge on a definite increase, an attendant increase is expected in the use of incineration for sludge disposal management. A characterization study was conducted on particulate emissions from the stacks of a group of municipal sludge incinerators, three with multiple-hearth furnaces and one with a fluidized-bed furnace. One purpose of the investigation was to provide information on the concentration of chemical elements, especially heavy metals such as cadmium (Cd) and lead (Pb), in the emissions. A related objective focused on obtaining source emission factors and source signatures to help develop and validate source apportionment models.

These investigations involved the collection of representative emission samples for: (1) particulate mass emission determination; (2) chemical characterization; and (3) particle size determinations, for both mass emissions and individual chemical element emissions. Sulfur dioxide and sulfuric acid emissions were determined; the gaseous concentrations of hydrocarbons and nitrogen oxides were monitored; and a few samples were collected to determine the emissions of high molecular weight organic compounds. At each incinerator, samples of the feed sludge were collected and analyzed. Information on operating conditions at the four incinerator facilities during the test periods are shown in Table 1.

At each incinerator, the sludge was preconditioned thermally with the hot exhaust gases from the furnaces to produce a higher solids sludge cake for better burning. At the multiple-hearth Incinerator O, this process was carried to the point of positive heat balance, resulting in autogenous burning. Some supplemental fuel was burned at all of the sites. At Incinerator O a total of only 16 gal of fuel oil was burned. This burning occurred during the final two 29-h sampling periods, and was carried out for shut-down purposes. The remainder of the time the

Table 1. *Furnace Types, Control Systems, and Sludge Loadings of Tested Incinerators*

Site designation	Furnace type	Control equipment	Average load of unit tested kg/h (dry)	Sludge percent moisture
O	Seven-chambered multiple-hearth	Wet, tray-type scrubber	1940	63.5
P	Eight-chambered multiple-hearth	Wet, tray-type scrubber	1740	49.3
Q	Fluidized-bed	Wet, tray-type scrubber	839	66.2
R	Seven-chambered multiple-hearth	Single pass cyclone scrubber	1890	71.3

sludge burned autogenously, combusting on its own with no need for supplemental fuel. At Incinerator P an average of 40 ft^3/h of natural gas was burned. The supplemental fuel oil at Incinerator Q ranged from 0 to 64 gal/h and averaged 17 gal/h over the three-day test period. Incinerator R was not designed for autogenous combustion and never reached this condition. An average of 40 gal/h of fuel oil was consumed.

Experimental

Sampling Procedures

A standard U.S. Environmental Protection Agency (EPA) Method 5 sampling train was used to collect particulate samples for determining mass emission rates. The impingers were adapted for use in determining gaseous sulfur oxides by EPA Method 8. The first impinger contained 150 mL of 80% isopropanol, which was followed by a high-purity glass wool plug inserted in the U-tube between the first and second impingers. The second and third impingers contained 100 mL each of 3% hydrogen peroxide solution; the fourth impinger was filled with approximately 400 g of indicating silica gel. Prior to sampling, a velocity profile of the duct at the sampling location was determined through transversing with a Pitot tube system. A 5-ft heated, Pyrex-lined probe was used to collect all Method 5 and Method 8 samples at a single point determined to have the average velocity of the flue gas within the duct.

Samples used for chemical characterization by x-ray fluorescence analysis (XRF) were collected with a modified Method 5 train in which the conventional sample box, filter holder, and glass impingers were replaced by an EPA-designed heated sample box housing a stainless steel filter holder for 47-mm filters. The particulate characterization samples were collected in sets consisting of two Gelman A glass fiber filters, two Millipore AA, one Nuclepore 0.8-μm filter, and six Teflon 0.2-μm filters. To provide a variety of loadings on the filters, sampling periods ranged from 15 sec to 10 min. In all cases, an attempt was made to sample isokinetically. Collections on all filters were analyzed by XRF.

University of Washington Mark III cascade impactors were used to collect samples for measuring the particle size distribution of total mass emissions, as well as individual chemical element emissions. The impactor samples were taken in-stack at the point of average gas velocity; this same sampling point was used for the characterization sampling. Samples were collected isokinetically, with a sampling rate between 0.5 and 0.75 ft^3/min through the impactor.

Composite pre-burn sludge samples and post-burn ash were collected at all incinerators except at the fluidized-bed unit, where only sludge samples were taken. The sludge samples were heated to 600°C for 30 min to remove the volatile contents prior to chemical analysis.

Analytical Methods

Elemental contents of the emission samples, as well as of the sludge and ash samples, were determined with a Siemens MRS-3 multi-channel wavelength dispersive x-ray fluorescence spectrometer. The Siemens MRS-3 has 15 fixed wavelength monochromators and a scanning channel that allows analysis for 11 additional elements.

Results and Discussion

Sludge Content

The elemental composition of the ashed sludge feed material, determined by XRF, is shown in Table 2. The values represent the averages for four to six composite samples taken during two- to three-day testing periods at each incinerator.

Elemental Emissions

The mean concentrations of the elements, determined by XRF, in the emissions from the four plants are shown in Table 3. The

Table 2. *Elemental Composition (Percent) of Municipal Wastewater Sludge*

Element	Site O		Site P		Site Q		Site R		Avg. Content 16 cities (a)
	AV	SD	AV	SD	AV	SD	AV	SD	
Na	1.0	0.09	0.7	0 11	<0.6		<0.2		0.44
Mg	1.8	0.08	1.0	0.17	0.87	0.13	0.92	0.03	0.60
Al	3 4	0.28	3.6	0 47	4.0	0.23	5.2	0.14	1.83
Si	10.1	0.46	13.0	3.3	17 0	2.7	12.0	0.34	—
P	6 4	0.44	4.4	0.22	4.1	0.13	3.7	0.12	1.56
S	0.93	0.15	1 3	0.41	0.82	0 46	0 47	0.06	—
Cl	0.45	0.01	0 08	0.18	<0.5		0.68	0.19	0 38
K	1 3	0.09	1.1	0 24	1.2	0 15	1.1	0.02	1 22
Ca	17.	0.62	6.2	1.2	7.5	1.0	9.8	0 15	3.62
Ti	0 84	0.06	0.76	0.05	0.77	0.04	1.18	0.04	0 23
V	0.03	0.01	0.04	0 01	0.03	0.01	<0.01		0.004
Cr	0.22	0.02	0.50	0.27	0.22	0.24	0.38	0.02	0.14
Mn	0 70	0.11	0.23	0 11	0.34	0.12	0.11	0 008	0.019
Fe	6 7	1.0	17	7 9	7.6	6.4	5.4	0 10	3.06
Co	0.01	0.003	0.03	0 02	0.01	0.02	0.01	0.002	0 001
Ni	0 03	0.01	0.14	0.08	0.06	0.07	0.06	0 008	—
Cu	0 12	0.02	0.75	0 43	0.27	0.37	0.44	0 02	0.13
Zn	0.30	0.04	1.1	0 08	1.0	0.07	1.4	0 03	0.21
As	<0 3		<0.3		<0.03		<0.03		0.0014
Se	<0.04		<0 04		<0.04		<0.04		0.0003
Br	<0 05		<0.05		<0.05		<0.05		0.005
Cd	0 02	0.005	0.04	0.05	0.08	0.04	0 03	0.005	0.010
Sn	0.30	0.02	0.25	0.03	0.25	0.03	0.19	0.006	0.022
Sb	0.03	—	0.02	0.004	0.01	0.001	0 01	0.001	0.001
Ba	0 25	0.02	0.41	0 11	0.45	0.04	0 27	0.01	0.06
Pb	0 34	0.13	0.48	0.18	0.59	0.25	0 23	0.03	0.18

(a) Furr, A.K., A.W. Lawrence, S S C. Tong, M.S. Grandolpho, R.A. Hofstader, C.A. Bache, W.H. Gutenmann, and D J. Lisk, Environ. Sci. Technol., 10,683, 1976

Table 3. *Mean Concentrations of Elemental Emissions (μg/Nm³) from Municipal Sludge Incinerators*

	Site O		Site P		Site Q		Site R	
Element	AV	SD	AV	SD	AV	SD	AV	SD
Na	350	110	290	230	100	50	240	170
Mg	90	32	440	550	51	18	21	6
Al	128	47	1570	1980	160	48	37	20
Si	590	100	4180	4826	270	80	620	290
P	590	120	1730	1890	210	69	570	350
S	660	300	930	410	730	270	2610	910
Cl	230	90	130	110	47	65	1990	2280
K	210	90	460	500	54	22	120	110
Ca	780	170	2620	2660	440	150	160	88
Ti	28	12	450	460	33	10	16	21
V	~35		38	14	9	2	<6	
Cr	97	39	480	330	14	8	230	106
Mn	27	13	82	30	23	8	10	1
Fe	370	80	7070	8170	228	71	230	145
Co	<6		25	8	<6		<6	
Ni	<22		125	121	<22		<22	
Cu	85	29	810	690	14	4	520	310
Zn	810	220	1840	1240	87	28	1830	1650
As	<29		<29		<29		<29	
Se	~45		20	3	18	17	26	1
Br	~49		57	19	39	17	170	140
Cd	42	15	34	15	7	2	1890	1410
Sn	180	51	1230	450	30	9	790	640
Sb	6	2	23	7	<2		43	40
Ba	10	4	290	280	20	7	14	15
Pb	510	190	1170	530	114	65	2140	1880

relative standard deviation of the samples analyzed (between 45 and 95 samples per site) to obtain each of these concentrations averaged 33%, 72%, 32% and 68% for O, P, Q and R, respectively. The greater variability of P and R reflect the wider range of operating conditions, e.g., the rates of feed and stack gas flow during the testing periods. It is apparent immediately that elements such as Zn, Cd, and Pb had a higher concentration in the emissions than would have been expected based solely on their concentration in the sludge. Enrichment factors, calculated by dividing the concentration in the total particulate emission by the concentration in the sludge, are listed in Table 4. Conditions causing enrichment are volatility and interaction of the volatile elements with the extremely fine particles not efficiently removed by scrubbers.

Total Particulate Mass and Sulfur Oxides

The emission rates and concentrations of particulate mass, sulfur dioxide and sulfuric acid were obtained from the Method 5 and Method 8 tests run after the scrubber outlets. Lowest particulate emissions were observed at the fluidized-bed incinerator, Q. Highest sulfur oxide emissions were found at Incinerator R, presumably due to the supplementary fuel oil burned to maintain combustion.

Particle Size Distribution

The average mass median diameters obtained from the particle size distribution measurements were 0.28, 0.30, 1.1 and 0.85 μm at Incinerators O, P, Q and R, respectively. Individual size distributions for several elements, as well as the total mass distribution, are shown in Figure 1. For convenience, the largest and smallest size fractions have been assigned finite values. The Zn distribution, predominantly submicrometer particles, was characteristic of that of S, Cd, Pb, and other elements exhibiting enrichment in the emissions, probably due to their volatility in the furnace. Phosphorus exhibited a group of mid-range particles around 2 μm. Iron was distributed through all sizes, with a significant fraction of large particles evident even in the controlled, post-scrubber emissions. Only a small fraction of the total mass was greater than 2 μm (see Figure 1).

At Incinerator R particle size measurements were also made before the scrubber. Figure 2 shows the distribution at the scrubber inlet, where large particles were preponderant; however, a second mode occurred near 2 μm and at the scrubber outlet, where the mass concentration of the smaller particles predominated because of the scrubber's more efficient removal of larger particles. Evident in these pre-scrubber measurement is the contrast in the distribution of a more volatile element such as Cd with that of calcium (Ca) (see Figure 2).

Conclusions

The more volatile elements, Cd, Pb and Zn, were enriched in the emissions. Cadmium had the highest average enrichment ratio, with a 31-fold increase in the emissions. The average mass median diameters for the particles emitted at all four sites were small, ranging from 0.28 μm to 1.1 μm. Few

Table 4. *Enrichment Ratios of Elements in Emissions Relative to Content in Sludge*

	Incinerator				Average
Element	O	P	Q	R	enrichment
Na	2.76	0.81	—	—	—
Mg	0.42	0.86	1.16	0.03	0.38
Al	0.30	0.85	0.78	0.01	0.49
Si	0.46	0.63	0.31	0.08	0.37
P	0.73	0.76	1.01	0.23	0.68
S	5.64	1.38	17.6	8.14	8.19
Cl	4.00	0.63	—	4.28	2.97
K	1.32	0.81	0.89	0.16	0.80
Ca	0.36	0.82	1.16	0.023	0.59
Ti	0.26	1.16	0.84	0.02	0.62
V	9.33	1.75	6.0	—	5.69
Cr	3.50	1.88	1.27	0.87	1.88
Mn	0.30	0.70	1.35	0.10	0.61
Fe	0.43	0.81	0.59	0.06	0.54
Co	—	1.67	—	—	—
Ni	—	1.71	—	—	—
Cu	5.67	2.11	7.0	1.75	4.13
Zn	21.47	3.25	28.7	1.89	13.83
Br	—	—	7.7	—	—
Cd	16.50	7.0	1.6	98.9	31.
Sn	4.73	9.56	2.36	6.10	5.68
Sb	1.67	2.50	—	4.6	2.92
Ba	0.32	1.36	0.89	0.07	0.66
Pb	11.97	4.70	3.83	13.65	8.53

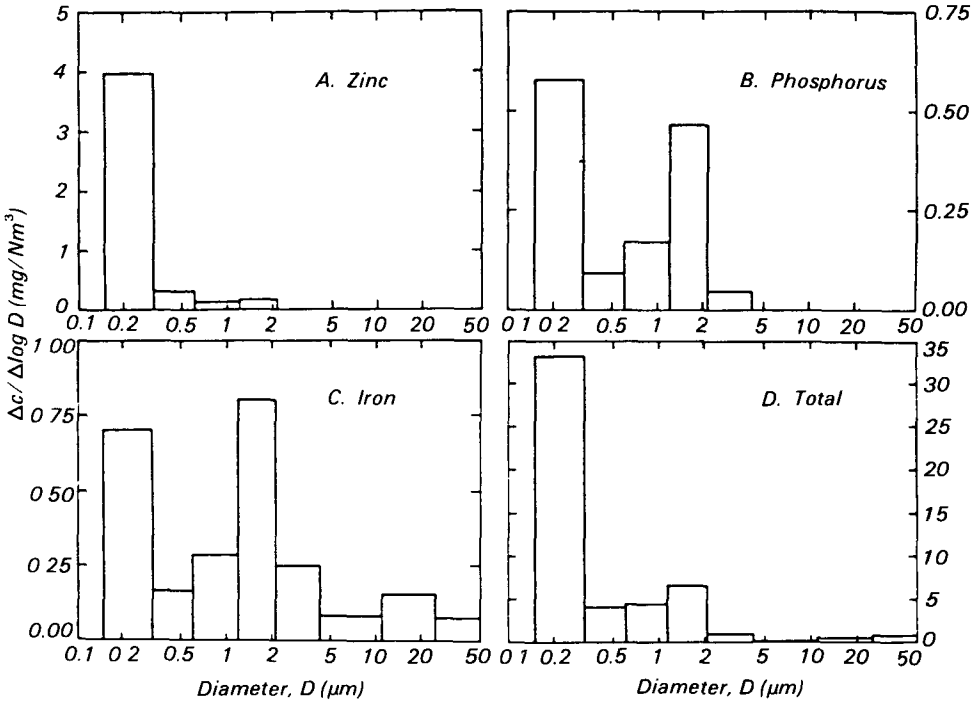

Figure 1. *Particle size distribution of zinc, phosphorus, iron, and total particulate emissions from Incinerator O.*

particles were larger than 2 μm. The volatile elements were found predominantly in the submicrometer range.

The enrichment of Cd (especially relative to that of Zn) is much greater at Incinerator R than at the other facilities. A contributing factor to this higher enrichment is Cd's greater predominance in small particles, as well as the lower efficiency of the control device. Incinerator R has a wet, single-pass scrubber system that is considerably less efficient for submicrometer particle removal than scrubbers at the other incinerators. Elemental particle size measurements at Incinerator R before the scrubber inlet showed that 72% of the Cd was associated with particles smaller than 1 μm (see Figure 2). Only 28% of the Zn was associated with this size range.

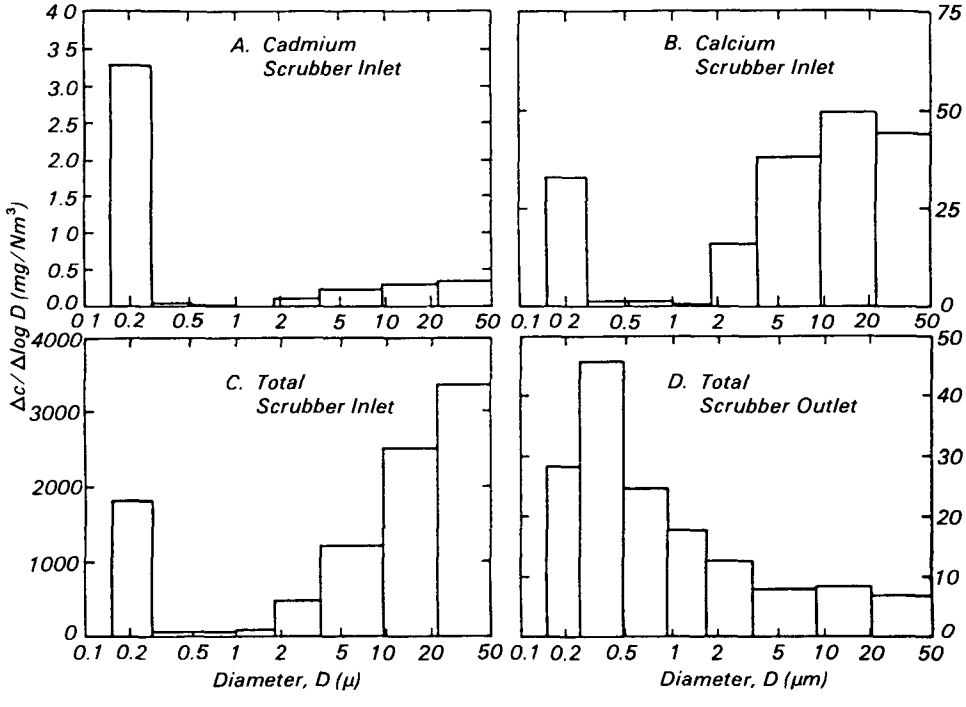

Figure 2. *Particle size distribution of cadmium, calcium, and total particulate mass before the scrubber and total particulate emission after the scrubber at Incinerator R.*

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The complete report, entitled "Chemical and Physical Characterization of Municipal Sludge Incinerator Emissions," (Order No. PB 84-169 325; Cost: $10.00, subject to change) will be available only from:

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