EPA/600/A-92/079

INCINERATION DATA ON ARSENIC AND LEAD EMISSIONS

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ABSTRACT

Since 1988, nine test programs have been conducted at the Environmental Protection Agency's Incineration Research Facility aimed at evaluating the fate of trace metals in the rotary kiln incineration of hazardous wastes and Superfund site materials. Results of six of those test programs have been reported to date. Of these six, two were parametric test series using a synthetic hazardous waste formulation and four were incineration treatability test programs using contaminated Superfund site materials. Results of these six test programs show remarkably consistent arsenic and lead partitioning behavior among the incinerator system discharge streams.

Overall test programs lead exhibits relatively nonvolatile behavior over a kiln temperature range from nominally 815°C (1,500°F) to 980°C (1,800°F) provided no chlorine is present in the feed material. Arsenic also exhibits relatively nonvolatile behavior over the same temperature range regardless of whether the feed contains chlorine at levels up to nominally 8 percent. Arsenic may be more volatile in the incineration of environmental samples such as Superfund site wastes than it is from a synthetic waste in which arsenic is introduced as As_2O_3 in aqueous solution. However, even with environmental samples, behavior is relatively nonvolatile. Lead volatility significantly increases at all kiln temperatures as feed chlorine content increases from 0 to as high as 8 percent.

INTRODUCTION

In 1988, the EPA's Risk Reduction Engineering Laboratory initiated a research program at its Incineration Research Facility (IRF) in Jefferson, Arkansas, to investigate the fate of trace metals fed to a rotary kiln incinerator. The initial test program was a five-test parametric study of the fate of five hazardous constituent trace metals (arsenic, barium, cadmium, chromium, and lead) and four nonhazardous constituent trace metals (bismuth, copper, magnesium, and strontium). In the parametric study each metal's partitioning to the incinerator's discharge streams (kiln ash, wet scrubber air pollution control system scrubber liquor, and flue gas) was measured, and the effects of kiln temperature, afterburner temperature, and feed chlorine content on metal partitioning were evaluated. The parametric tests were performed with a synthetic waste feed mixture prepared by adding a mixture of organic compounds (toluene, chlorobenzene, and tetrachloroethene) to a clay-based oil sorbent material, which was screw fed to the incinerator. The test metals were added by metering a concentrated aqueous solution containing the metal into the clay/organic mixture in the screw feeder.

The first parametric study, completed in 1988, investigated a venturi scrubber, packed-column scrubber combination for particulate and acid gas control. A second parametric study, identical in scope to the first, was completed in 1989. The only difference between the first and second studies was the air pollution control system (APCS), which was a single-stage ionizing wet scrubber. Results of the studies were reported in detail in 1991.

A third parametric study was completed in 1991. This study added mercury to the set of test metals and used a Calvert Force/Flux Condensation scrubber system as the APCS. Results of this study are only now being evaluated.

Shortly after beginning the trace metal fate research program at the IRF, requests to provide pilot-scale incineration treatability data to support EPA Regional Office-administered Superfund site remedial actions began to come to the IRF. Many of the contaminated materials at these sites contain not only organic contaminants, but hazardous constituent trace metals as well. Thus, key questions often asked by the Regions concerned the fate of contaminant trace metals, often as affected by incinerator operation.

As the result of the combination of a series of parametric metals partitioning studies using synthetic hazardous waste material with a series of Superfund site metals-contaminated materials tests, the research program at the IRF has generated a substantial body of metals partitioning data over the past 3 years. Somewhat surprisingly, the metals partitioning data from the wide variety of feed materials tested has been quite consistent.

This paper looks at the body of arsenic and lead distribution data to give some examples of the general conclusions that have resulted from metals partitioning research program at the IRF. Arsenic and lead were chosen for two reasons. They are the most common metal contaminants in the Superfund site contaminated materials tested at the IRF. In addition, the arsenic behavior seen is not what might have been expected a few years ago, and the lead partitioning behavior can be dramatically altered by incineration temperature and waste feed composition.

TEST PROGRAMS

Test Facility

All test programs discussed in this paper were performed in the IRF's rotary kiln incinerator system (RKS). A process schematic of the RKS is shown in Figure 1. The IRF RKS consists of a primary combustion chamber, a transition section, and a fired afterburner chamber. After exiting the afterburner, flue gas flows through a quench section followed by a primary air pollution control system (APCS). Two primary APCSs are available for use on the unit, as shown in Figure 1. One consists of a venturi scrubber followed by a packed-column scrubber fabricated by Andersen 2000. The other is a single-stage ionizing wet scrubber fabricated by Air Plastics, Inc. Downstream of the primary APCS, a backup secondary APCS, comprised of a demister, an activated-carbon adsorber, and a high-efficiency particulate (HEPA) filter, is in place. This secondary APCS is designed to ensure that particulate and organic emissions from the system are acceptable even under upset conditions.

Test Waste Materials

Results from six different test programs are discussed in this paper. As noted in the Introduction, two test programs were the parametric trace metal fate programs performed in 1988 and 1989 using a synthetic hazardous waste feed and employing the venturi/packed-column scrubber for one program and the ionizing wet scrubber for the other. The Introduction also noted that the synthetic waste feed materials for the parametric test programs was prepared by combining a mixture of toluene, with varying amounts of chlorobenzene and tetrachloroethene with a clay-based oil sorbent.

The clay/organic mixture contained nominally 30 percent by weight organic liquids, though it remained a free-flowing solid. Test trace metals were added to the clay/organic mixture by metering a concentrated aqueous metals solution onto the clay/organic mixture at the head of the screw feeder used to feed the synthetic waste to the kiln. Lead in the form of its soluble nitrate ($Pb(NO_3)_2$) at 1.6 to 3.2 g/L lead, and arsenic as dissolved As_2O_3 also at 1.6 to 3.2 g/L arsenic were contained in the concentrated aqueous solution.

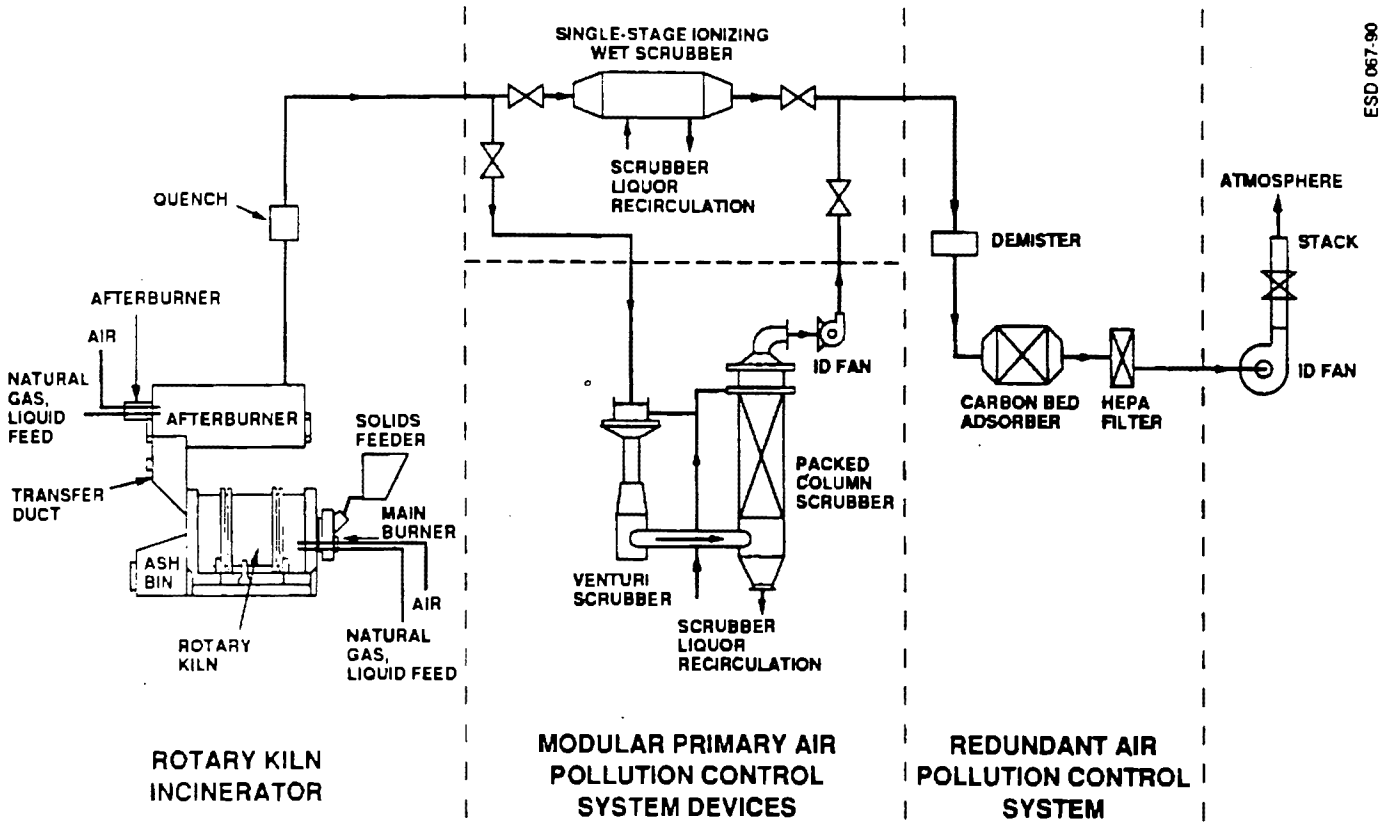

Figure 1. Schematic of the IRF rotary kiln incineration system.

In addition to the synthetic waste parametric tests, contaminated materials from four Superfund sites were tested at the IRF kin the late 1989 to early 1991 time frame. These materials were arsenic and/or lead-contaminate, and one test objective was to evaluate trace metal fate. The Superfund site test programs included:

- Three lead-contaminated materials from the waste pit area of the Purity Oil Sales Superfund site near Fresno, California. The three materials were the top layer of the waste pit (the A layer), composed of contaminated construction rubble, sand, and gravel; and underlying tar sludge mixed with some soil and rubble (the B layer); and a base layer of contaminated silty sand (the C layer).
- Pesticide-, arsenic-, and lead-contaminated soil from the Baird and McGuire Superfund site in Holbrook, Massachusetts
- PCB-contaminated marine sediments dredged from the Hot Spot Operable Unit of the New Bedford Harbor Superfund site near New Bedford, Massachusetts. The sediments were also contaminated with over 200 mg/kg lead.
- An arsenic- and lead-contaminated soil from the Drake Chemical Superfund site in Lock Haven, Pennsylvania

Table I summarizes the arsenic and lead concentrations in the synthetic waste feeds used in the parametric trace metal fate test programs and in the six Superfund site materials from four sites tested in the Superfund site materials tests. Also shown in Table I is the chlorine content of each test material. The importance of feed chlorine contents is discussed in the Test Results section of this paper.

Table I. Test feed material arsenic, lead, and chlorine content.

Test feed	Concentration, as received/prepared		
	Arsenic, mg/kg	Lead, mg/kg	Chlorine, %
Venturi scrubber parametric test synthetic waste			
No chlorine	25	33	0
Low chlorine (average)	24	29	4.0
High chlorine	33	32	8.3
Ionizing wet scrubber parametric test synthetic waste			
No chlorine	41	39	0
Low chlorine (average)	51	47	3.5
High chlorine	52	47	6.9
Purity Oil sales site contaminated materials			
A layer	5	860	<0.2
B layer	4	10,200	<0.3
C layer	<4	780	<0.2
Baird and McGuire site contaminated soil (average)	85	20	<0.2
New Bedford Harbor site contaminated sediments (composite)	NA[a]	236	0.85
Drake Chemical site M-2 soil	16	439	<0.2

[a]NA = not analyzed.

Test Conditions

The parametric trace metal fate tests were specifically designed to evaluate whether metal partitioning was affected by incinerator operation and feed composition. The feed composition variable chosen for evaluation was chlorine content, as noted in Table I. The key incinerator operating variable chosen was kiln temperature. Similarly, the for Baird and McGuire site soil tests, EPA Region 2 was interested in whether kiln temperature affected arsenic partitioning, and for the New Bedford Harbor site sediment tests, the Region was interested in whether kiln temperature affected PCB destruction and lead partitioning. All three Purity Oil Sales site materials and the Drake Chemical site soil were tested under a single set of nominal incinerator operating conditions.

Table II summarizes the incinerator operating conditions for each of the tests. As shown in the table, kiln temperature variations tested ranged from nominally 815°C (1,500°F) to 980°C (1,800°F). All tests except the Purity Oil Sales site tests and the New Bedford Harbor site tests were performed at a nominal afterburner temperature of 1,090°C (2,000°F). The Purity Oil Sales site tests were performed with nominal afterburner temperature of 980°C (1,800°F) and the New Bedford Harbor site tests were performed with nominal afterburner temperature of 1,200°C (2,200°F).

Table II. Incinerator operating conditions.

Test	Kiln exit		Afterburner exit	
	Temperature, °C (°F)	Flue gas O_2, %	Temperature, °C (°F)	Flue gas O_2, %
Venturi scrubber parametric tests				
Low kiln temperature, low chlorine	825 (1517)	11.9	1071 (1959)	8.4
Moderate kiln temperature				
No chlorine	874 (1606)	12.2	1093 (2000)	7.1
Low chlorine	878 (1612)	12.4	1088 (1991)	7.9
Duplicate test	873 (1603)	11.4	1093 (2000)	7.6
High chlorine	870 (1599)	11.4	1092 (1998)	7.4
High kiln temperature, low chlorine	928 (1701)	10.6	1092 (1998)	7.1
Ionizing wet scrubber parametric tests				
Low kiln temperature, low chlorine	819 (1507)	12.3	1095 (2002)	7.7
Moderate kiln temperature				
No chlorine	900 (1652)	12.0	1088 (1990)	8.3
Low chlorine	877 (1610)	11.9	1096 (2006)	7.3
Duplicate test	881 (1618)	12.6	1103 (2018)	7.6
Triplicate test	879 (1615)	12.6	1098 (2008)	8.1
High chlorine	881 (1617)	12.9	1087 (1988)	8.1
High kiln temperature, low chlorine	929 (1704)	11.9	1092 (1998)	7.3
Purity Oil sales site tests				
A layer	875 (1607)	12.6	980 (1796)	10.5
B layer	881 (1617)	11.6	984 (1804)	9.9
C layer	827 (1611)	12.3	1001 (1834)	10.0
Baird and McGuire site tests				
Low kiln temperature, low kiln O_2	844 (1552)	6.8	1089 (1993)	6.3
Low kiln temperature, high kiln O_2	831 (1529)	11.4	1094 (2002)	7.9
Duplicate test	838 (1541)	11.3	1083 (1981)	8.1
High kiln temperature, low kiln O_2	994 (1822)	7.5	1099 (2009)	7.3
High kiln temperature, high kiln O_2	994 (1822)	10.4	1105 (2021)	7.4
New Bedford Harbor site tests				
Low kiln temperature	824 (1516)	11.2	1208 (2206)	6.4
High kiln temperature	984 (1803)	9.0	1208 (2206)	6.0
Duplicate test	985 (1805)	10.0	1208 (2206)	7.0
Drake Chemical site test				
Soil M-2	826 (1519)	13.3	1096 (2005)	8.7

Sampling and Analysis

For all tests, concentrations of the trace metals of interest were measured in the incinerator feed, the kiln ash discharge, the scrubber liquor, and the scrubber exit flue gas. Flue gas samples for metals analysis were collected using the multiple metals sampling train described in the hazardous waste incinerator guidance document (Reference 1) for all tests except the Purity Oil Sales site tests. An EPA Method 12 sampling train (Reference 2) was used for lead (the only test flue gas metal of interest) in these tests.

Arsenic and lead analyses of test samples was by inductively coupled argon plasma (ICAP) spectroscopy (EPA Method 6010, Reference 3) for all tests except arsenic for the Drake Chemical site tests and both metals for the parametric tests using the venturi/packed-column scrubber. Graphite furnace atomic absorption (GFAA) spectroscopy methods were used instead for these exceptions (Method 7060 for arsenic and 7421 for lead.

TEST RESULTS

The measured feed and discharge stream arsenic and lead concentrations can be combined with measured feed and discharge flowrates, and the fraction of both metals fed accounted for the respective discharges can be calculated. The sum of these discharge fractions represents the mass balance closure for each metal in each test. Ideally, near 100 percent trace metal mass balance closure would be desirable. However, past experience in tests to determine the distribution of trace metals from combustion sources has shown that typical good results are in the 30 to 200 percent range. For the tests discussed in this paper, lead mass balance closures range from 8 to 177 percent and averaged 92 percent. Arsenic closures ranged from 32 to 101 percent and averaged 67 percent.

Given that variable and less than perfect mass balance closure is invariably experienced, it is difficult to draw conclusions regarding the affect of incinerator operation or feed characteristics on metal partitioning using only percent-of-feed fractional distributions. However, a clearer picture of the variation in relative metal distributions is possible when percent-of-feed fractional distributions are normalized by the total mass balance closure achieved. These normalized, or percent-of-measured fractions represent fractions that would have resulted had mass balance closure in each case been 100 percent. Use of distribution fractions normalized in this manner allows clearer data interpretation, because variable mass balance closure is removed as a source of test-to-test data variability. The use of normalized distributions represents a best attempt to quantify metal partitioning phenomena, given variable and less than perfect mass balance closure.

Table III summarizes the normalized distribution fraction of arsenic and lead measured in each of the tests listed in Table II. For completeness, the total mass balance closure achieved for each metal (ratio of the sum of amount of each metal measured in the discharges to the amount of metal fed to the incinerator) is also shown in Table III for each test.

The test condition data given in Table II show that over the six test programs performed, kiln temperature was varied over four nominal levels: 815°C (1,500°F), 870°C (1,600°F), 925°C (1,700°C), and 980°C (1,800°F). The feed composition data in Table I show that feed chlorine content also varied over four nominal levels; no chlorine, 1 percent, 4 percent, and 8 percent.

Figure 2 plots the normalized lead kiln ash fraction data from Table III in bar chart form for all the tests with no feed chlorine. For the purpose of the Figure 2 plot, the three Baird and McGuire site tests at the low kiln temperature and the two tests at the high kiln temperature were considered replicates. This presumes that the kiln exit flue gas O_2 variations tested had no effect on metal partitioning. This clearly appears to be the case for the lead distribution data in Table III. For these replicates, the height of the bar in Figure 2 represents the average of the replicate fractions. The range of normalized fractions measured over the replicates is shown by the "error bar" superimposed in the figure.

Table III. Normalized arsenic and lead discharge distributions.

Test	Normalized distribution, % of metal measured							
	Arsenic				Lead			
	Kiln ash	Scrubber liquor	Scrubber exit flue gas	Mass balance closure achieved	Kiln ash	Scrubber liquor	Scrubber exit flue gas	Mass balance closure achieved
Venturi scrubber parametric tests								
Low kiln temperature, low chlorine	94	3	3	76	13	37	50	90
Moderate kiln temperature								
No chlorine	94	4	2	68	84	5	11	8
Low chlorine	86	8	6	46	15	36	49	84
Duplicate test	92	4	4	39	14	36	50	37
High chlorine	92	3	5	52	6	20	74	96
High kiln temperature, low chlorine	84	8	8	49	11	22	67	77
Ionizing wet scrubber parametric tests								
Low kiln temperature, low chlorine	92	3	5	64	91	1	8	130
Moderate kiln temperature								
No chlorine	89	4	7	66	90	1	9	47
Low chlorine	80	9	11	47	81	7	12	136
Duplicate test	90	4	6	62	82	5	13	94
Triplicate test	89	5	6	77	85	4	11	102
High chlorine	95	3	2	65	83	10	7	95
High kiln temperature, low chlorine	82	11	7	95	71	14	15	177

Table III. Normalized arsenic and lead discharge distributions (concluded).

Test	Normalized distribution, % of metal measured							
	Arsenic				Lead			
	Kiln ash	Scrubber liquor	Scrubber exit flue gas	Mass balance closure achieved	Kiln ash	Scrubber liquor	Scrubber exit flue gas	Mass balance closure achieved
Purity Oil sales site tests								
A layer					94	1	5	155
B layer					91	2	7	126
C layer					96	1	3	169
Baird and McGuire site tests								
Low kiln temperature, low O_2	76	22	2	101	93	3	4	148
Low kiln temperature, high O_2	72	23	5	82	89	4	7	113
Duplicate test	66	29	5	74	91	3	6	98
High kiln temperature, low O_2	56	38	6	78	69	13	18	51
High kiln temperature, high O_2	36	55	9	62	69	12	19	37
New Bedford Harbor site tests								
Low kiln temperature					53	30	17	66
High kiln temperature					23	35	42	38
Duplicate test					19	26	55	54
Drake Chemical site tests								
Soil M-2	>77	<8	15	73	90	8	2	73

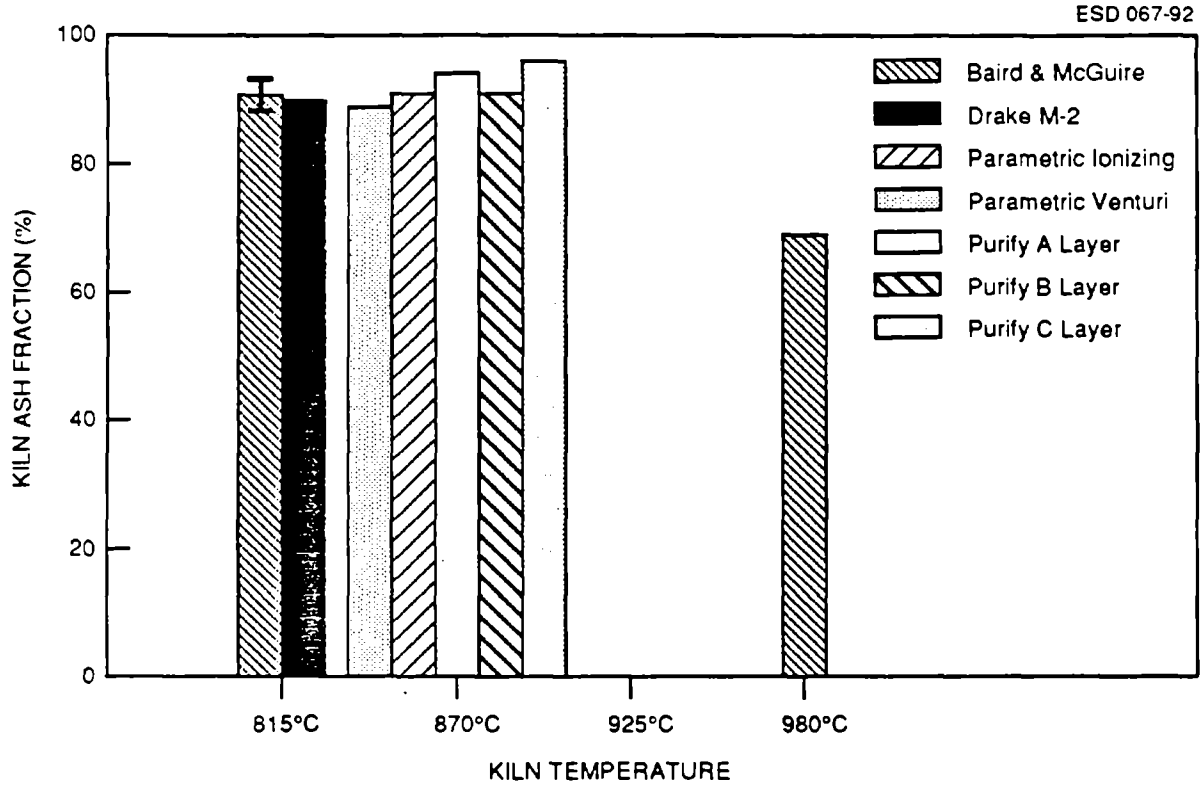

Figure 2. Normalized kiln ash fractions for lead; tests with no feed chlorine.

The data in Figure 2 show that lead was relatively nonvolatile at a kiln temperature of nominally 815°C (1,500°F). For the two different Superfund site materials tested at this nominal temperature, about 90 percent of the lead discharge was accounted for in the kiln ash.

Similar kiln ash fractions were measured for the five materials tested at a nominal kiln temperature of 870°C (1,600°F). However, lead became much more volatile at a nominal kiln temperature of 980°C (1,800°F). At this temperature for the Baird and McGuire soil, the kiln ash accounted for a significantly reduced fraction, about 70 percent of the lead discharged.

Similar behavior is seen in Figure 3, which is a bar chart plot of the ionizing wet scrubber parametric test data at low feed (4 percent) chlorine content and the New Bedford Harbor data (sediments contained roughly 1 percent chlorine). For the nominal 870°C (1,600°F) ionizing wet scrubber test conditions, the height of the bar represents the average of the three replicate test kiln ash fractions from Table III. The kiln ash fraction range over the three replicate tests is shown by the superimposed error bar.

Figure 3 shows a steady increase in lead volatility as measured by steady decrease in lead kiln ash fraction over the nominal 815° to 925°C (1,500° to 1,700°F) range for the ionizing wet scrubber parametric tests. Lead exhibited significantly more volatile behavior in the New Bedford Harbor sediment tests. Only about 50 percent of the lead discharged was accounted for in the kiln ash for the test at nominal kiln temperature of 815°C (1,500°F). The kiln ash fraction decreased to about 20 percent at the higher kiln temperature of nominally 980°C (1,800°F).

Arsenic shows similar behavior as illustrated in Figure 4 which is a bar chart plot of arsenic kiln ash fractions for all tests with arsenic present in the feed. For both sets of synthetic waste parametric tests, arsenic kiln ash fractions steadily decreased from just over 90 percent at a nominal 815°C (1,500°F) kiln temperature to just at 90 percent at a nominal 860°C (1,600°F) kiln temperature, to just under 85 percent at a nominal 925°C (1,700°F) kiln temperature. Arsenic was

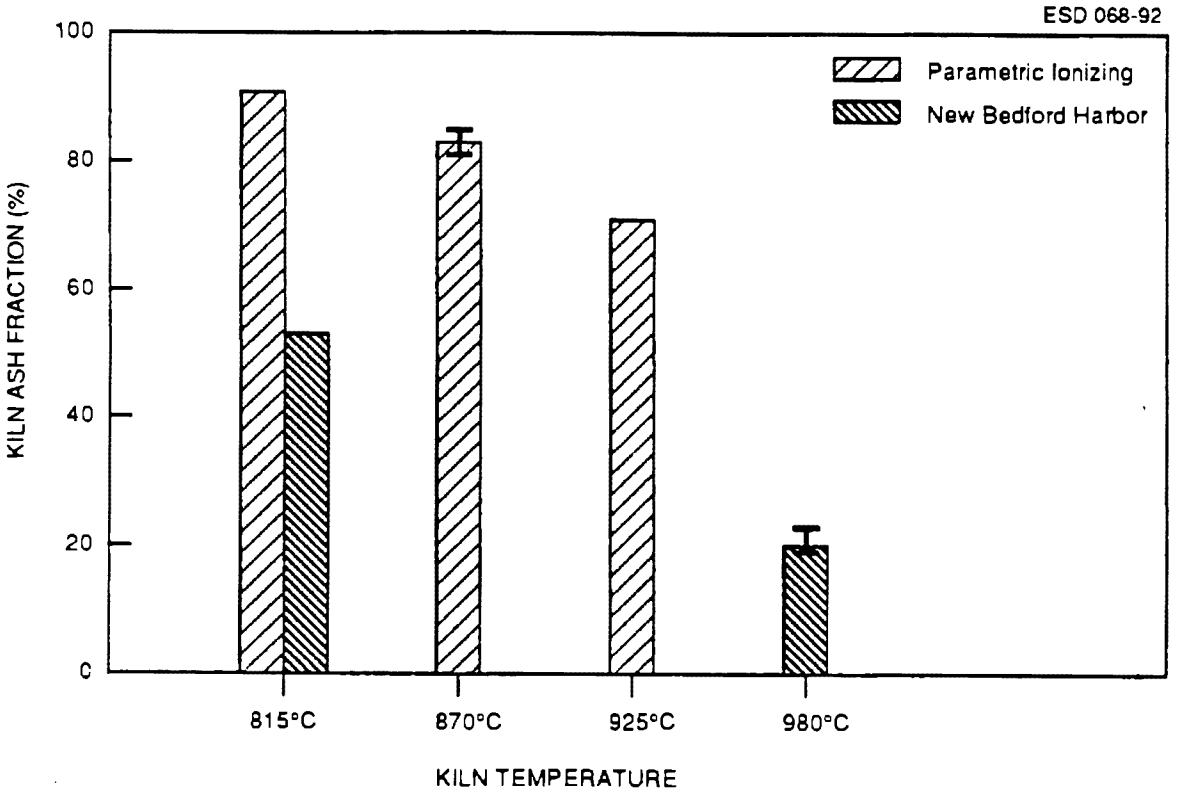

Figure 3. Normalized kiln ash fractions for lead; tests with low (1 to 4 percent) feed chlorine.

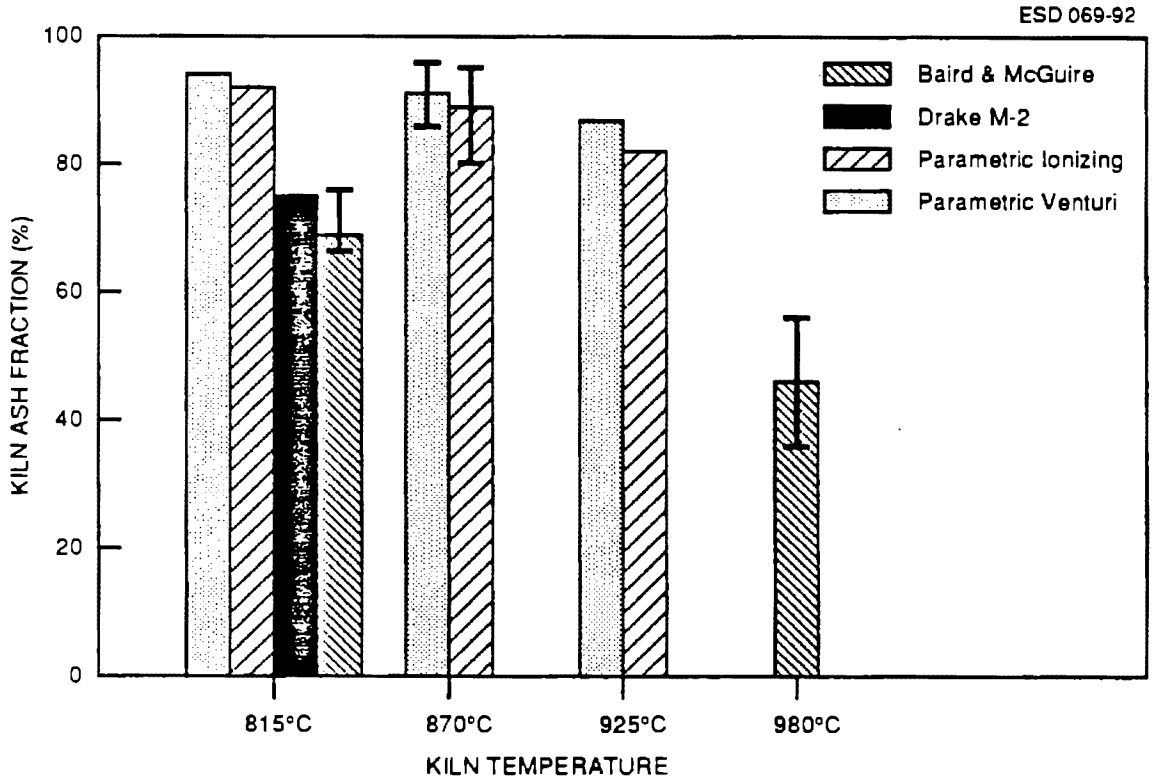

Figure 4. Normalized kiln ash fractions for arsenic.

more volatile at 815°C (1,500°F) from the Drake Chemical site soil, and even more volatile at this temperature from the Baird and McGuire site soil. However, the kiln ash still accounted for just over 70 percent of the arsenic discharged. Arsenic exhibited significantly more volatile behavior from the Baird and McGuire soil at the high kiln temperature condition of about 980°C (1,800°F). However, even at this temperature the kiln ash still accounted for just under half the arsenic discharged.

The effect of feed chlorine content on lead volatility is shown in Figure 5. For all tests in the base discussed in this paper, the presence of even a small amount of feed chlorine (i.e., 1 percent) caused a significant increase in lead volatility as measured by a decrease in normalized lead kiln ash fraction. The parametric tests with synthetic waste at a nominal kiln temperature of 870°C (1,600°F) show a decrease in lead kiln ash fraction as feed chlorine content is increased from 0 to nominally 4 and 8 percent. The venturi scrubber parametric test data show a dramatic increase in lead volatility with increasing feed chlorine content. The ionizing wet scrubber parametric show a less prominent volatility increase with increased feed chlorine content to 4 percent, but a definite one nonetheless.

Even the data from three Superfund site materials tests supports the observation that the presence of feed chlorine increases the volatility of lead. Lead was relatively nonvolatile from both the Drake Chemical site M-2 soil and the Baird and McGuire site soil tested at a kiln temperature of nominally 815°C (1,500°F). However, lead was significantly more volatile from the New Bedford Harbor sediments, which contained about 1 percent chlorine, at this temperature. The same relative behavior is seen for the Baird and McGuire site and New Bedford Harbor site tests at a kiln temperature of nominally 980°C (1,800°F).

CONCLUSIONS

The results from six test programs completed at EPA's IRF over the past three years show remarkably consistent partitioning behavior of arsenic and lead in rotary kiln incinerator discharge streams. This consistent behavior exists over a range of test feed materials from a synthetic clay-based sorbent material through contaminated soils, sludges, and marine sediments from four Superfund sites. Over all these tests:

- Lead exhibits relatively nonvolatile behavior over a kiln temperature range from nominally 815°C (1,500°F) to 980°C (1,800°F) provided no chlorine is present in the feed material. The kiln ash discharge accounts for about 90 percent of the lead measured in the discharge streams at kiln temperatures of 815°C and 870°C (1,500° and 1,600°F), decreasing to about 70 percent at a kiln temperature of 980°C (1,800°F).
- Arsenic exhibits relatively nonvolatile behavior over the same temperature range regardless of whether the feed contains chlorine at levels up to nominally 8 percent. For a synthetic waste feed which incorporates arsenic as As_2O_3 in aqueous solution, the kiln ash accounts for just over 90 percent of the arsenic measured in incinerator discharges at a kiln temperature of 815°C (1,500°F), decreasing to just under 85 percent at a kiln temperature of 925°C (1,700°F).
- Arsenic may be more volatile in the incineration of environmental samples such as Superfund site wastes. However, overall behavior is still relatively nonvolatile. the kiln ash accounted for at least 70 percent of the arsenic discharged in the incineration of Superfund site materials at a kiln temperature of 815°C (1,500°F) decreasing to just under 50 percent at a kiln temperature of 980°C (1,800°F).
- Lead volatility is significantly increased with chlorine present in the incinerator feed, with increased volatility exhibited as feed chlorine content progressively increases from 0 to 8 percent.

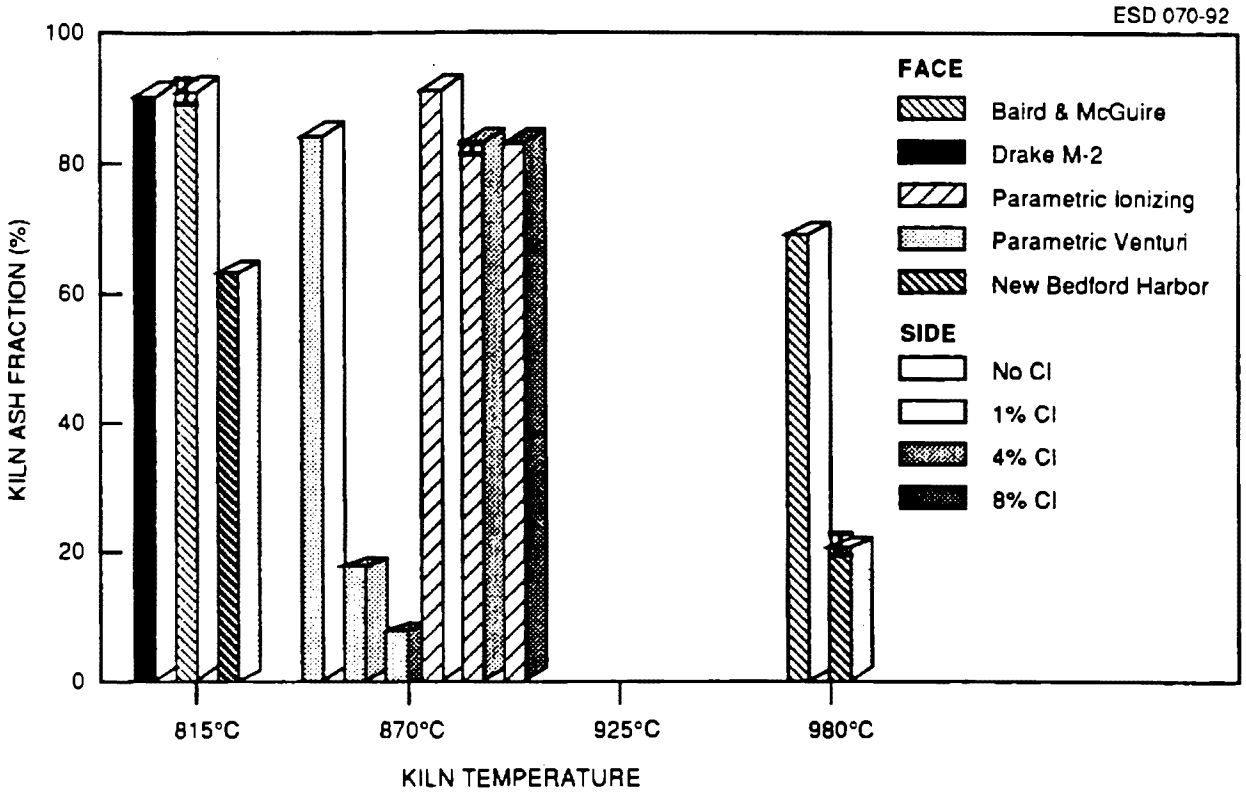

Figure 5. Normalized kiln ash fractions for lead; effect of feed chlorine.

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16. ABSTRACT

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