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Fate and behavior of selected heavy metals in incinerated sludge

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Since the early 1940s, it has become increasingly difficult to sell, give away, or dispose of raw or partially dewatered wastewater sludges. Conventional methods of handling, including ocean dumping, landfill disposal, lagooning, and placing on sand beds, have become costly and unviable in terms of legislation, health, and appearance. During the next 3-5 years, disposal problems will be further magnified by an anticipated doubling of sludge volumes. This is due to the expansion and construction of wastewater treatment plants, and the requirement to cease ocean dumping of harmful wastewater sludges by 1981.¹

In the New York-New Jersey metropolitan area, where 90% of the nation's ocean dumping problem exists, there has been renewed interest in sludge incineration, which can accomplish a maximum reduction of waste solids. Of concern, however, is the potential air pollution resulting from this combustion process, particularly the emission of heavy metals. To address this problem, field-scale mass balance studies were undertaken by Rutgers University at the fluidized bed sludge incinerator, operated by the Northwest Bergen Sewer Authority, Waldwick, N. J. Data on the fate of Cd, Cr, Cu, Ni, Pb, Zn, and Hg were developed.

FIELD-SCALE STUDIES CONDUCTED

The Authority's treatment facility services nine northern New Jersey suburban communities. Full activated sludge treatment is provided for the daily flow of 18 925 m³ (5.0 mil gal). Industrial contributions are estimated to be less than 10% of the total flow.

The fluidized bed sludge incinerator, rated at 499 kg/h (1 100 lb/h) on a dry solids basis, operates at a bed temperature of approximately 788°C (1 450°F).

Dewatered wastewater sludge at approximately 18% solids is fed into the fluidized bed, which is composed of graded silica sand. Air is supplied to the reactor as close to the fuel as is practical, and thoroughly mixed with the dewatered sludge so that combustion may be completed in a short time. Sufficient air is used to keep the sand in suspension and at the same time, prevent a carry over of a major portion of the sand from the reactor. The violent mixing of the solids and gases results in uniform conditions of temperature, composition, and particle size distribution throughout the bed. Heat transfer between the gases and solids is extremely rapid because of the large surface area available. The bed retains the organic sludge particles until they are reduced to mineral ash, which is constantly stripped from the reactor by the up-flowing gases.

It is noted that, depending on the design and operating characteristics, a portion of the bed sand, particularly the finer particles, is contained in the carry-over gases. Ash leaves the reactor with the combustion gases through a refractory lined duct, heat exchanger, contact venturi, and finally into a five-plate "Peabody Impingement" scrubber. Effluent from the secondary treatment process is used in the scrubber, and when separated from the ash, is directed back to the head of the treatment facility. Figure 1 illustrates the flow of gases, ash, and water through the reactor system.

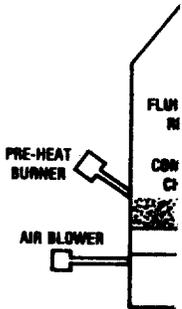


FIGURE 1. Fluidized bed sludge incinerator.

SAMPLING AND PROCEDURES

Three separate samplings were conducted during the study at the treatment facility. The sampling points are shown in Figure 2. Sludge, scrubber wastewater, and ash were sampled. They were composed of a balance from the cyclone separator, a recirculating plant, were accounted for, though not independent operating day.

Flow measurements were made at the time a sample was taken. Flows were measured with a flow watch, while effluent flows were measured with a flow meter, respectively. Totalizer readings were used to generate calibration curves. Calibration was done by plant personnel at the actual incinerator. Stack particulate sampling was done using the standard method of the Environmental Protection Agency (EPA).² Background measurements were made using the glass fiber filter method. The major portion of the calculations for each metal were done using the standard method of the Environmental Protection Agency (EPA).²

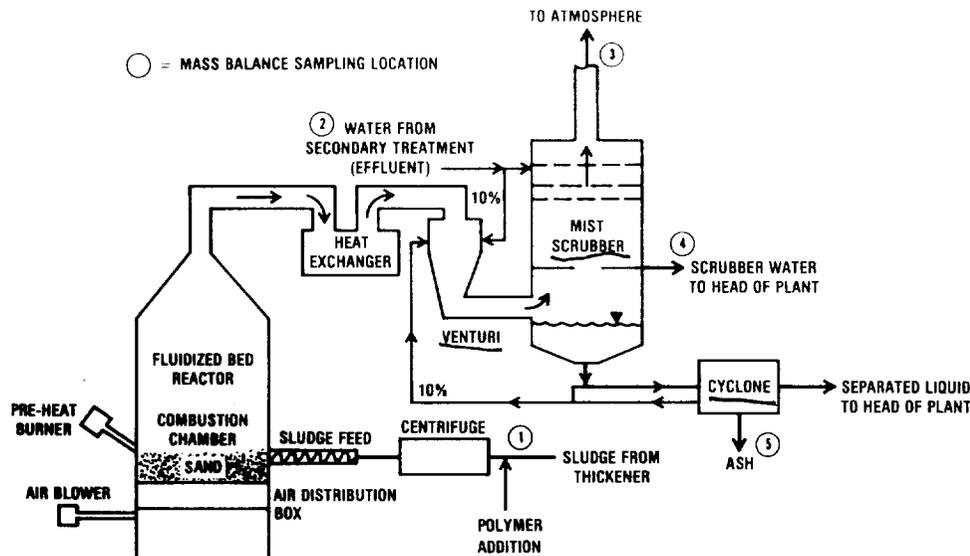


FIGURE 1. Fluidized bed wastewater sludge incinerator—Northwest Bergen.

SAMPLING AND ANALYTICAL PROCEDURES

Three separate heavy metal mass balances were conducted during a normal operating day at the treatment facility. Sampling locations are shown in Figure 1. Samples of the feed sludge, scrubber water, secondary (final) effluent, and ash were collected every half-hour. They were composited, based on flow, for each sampling or balance period. Separated liquids from the cyclone and centrifuge, which represent a recirculating load through the treatment plant, were accounted for in the balance, although not independently sampled during the operating day.

Flow measurements were also obtained each time a sample was collected. Sludge and ash flows were measured using a bucket and stopwatch, while effluent and scrubber water were measured, respectively, by using the plant's totalizer readings and pump characteristic curves. Calibration of both these systems was done by plant personnel 2 weeks prior to the actual incinerator test run.

Stack particulate emissions were obtained using the standard U. S. Environmental Protection Agency (EPA) sampling train, Method 5.² Background heavy metal concentrations in the glass fiber filters were determined prior to conducting the mass balance studies. Corrections for each metal were applied to the final calculations.

During collection of mercury samples, the two impingers following the filter contained 20 ml of KMnO_4 , 5 ml of mercury-free H_2SO_4 , and 2 ml of mercury-free HNO_3 , and were then brought up to a total volume of 75 ml with the addition of deionized water. This modification was intended to oxidize the captured organo-mercurials immediately because the possibility existed for the mercury entering the sampling train to pass through the glass fiber filter as vapor, and therefore escape detection. The total contents of the impingers, plus the filter, were then analyzed in accordance with EPA's recommended procedure for "Mercury in Sediment."³ The alternate digestion procedure, employing an autoclave, was followed.

A sampling traverse was not performed as specified in Method 5. Instead, a velocity traverse was completed in compliance with Methods 1 and 2 of the standards of performance.³ From these data, an average stack velocity was calculated. During sampling, a traverse point was selected at which the velocity was equal to the average velocity, and the entire 60-minute sample was withdrawn from this point under isokinetic conditions. Sampling was performed at approximately 4 m (13 ft) above the stack inlet to the scrubber.

All samples were iced (4°C) immediately following collection and returned to the laboratory for analysis within 6 hours. Sludge and

TABLE I. Plant operating and sludge characteristics: mass balance study Northwest Bergen fluidized bed incinerator.

Sampling Period	Sludge Feed Characteristics			Ash Characteristics			Scrubber		Stack		
	Solids, %	Volatiles, %	Flow, l (gal)	Input, g/period	Solids, %	Flow (Wet) g/min (lb/min)	Output, g/period	Flow, l/min (gal/min)	Temperature, °C (°F)	Flow, scm/min (dry) (scfm)	Velocity, m/min (ft/min)
A	1.60	81.8	35 954.5 (9 499.2)	575 478.2	57.5	562.45 (1.24)	77 742.50	1 325 (350)	26.7 (80)	99.3 (3 505)	11.0 (36.1)
B	1.65	82.9	28 208.8 (7 452.8)	465 613.2	57.3	630.49 (1.39)	66 843.95	1 325 (350)	26.7 (80)	103.9 (3 670)	11.6 (38.0)
C	1.60	81.7	17 768.3 (4 694.4)	284 370.8	51.7	630.49 (1.39)	44 019.27	1 325 (350)	26.7 (80)	109.5 (3 865)	12.2 (40.0)

* Measured after thickening and before centrifugation.

ash characteristics—percentage solids, moisture, volatiles, and pH—were determined immediately upon receipt in the laboratory, with the remainder of the sludge and ash samples being oven dried for 24 hours at 100°C in preparation for metals analyses. Ash samples for mercury analysis were dried at 60°C for 24 hours, and sludge samples were analyzed wet, to prevent possible loss of the mercury by air drying.

Trace metals analyses of the glass fiber filters, probe backwash, impinger contents, sludge ash, effluent, and scrubber water were performed by atomic absorption spectroscopy in accordance with EPA methodologies.³

All analyses were conducted at EPA's Region II Laboratory, Edison, N. J. Quality assurance procedures, which followed the everyday operating protocol of the EPA laboratory, involved approximately 12% of the samples collected, and included intralaboratory comparison of working standards, replication, split samples, standard addition, and maintenance of control charts for mean (\bar{X}) and range (R) for all metal analyses.

TRACE-METALS MASS BALANCE

Incinerator operating characteristics during the testing periods are summarized in Table I. Results of the three individual mass balance studies for all trace metals, except mercury, are shown in Table II. The "sampling day" summary, which represents the average of the 10.5 hours of testing, is shown in Table III.

Considering the potential for inorganic and organic mercury compounds to volatilize below 365°C (680°F), it was not unexpected during this study, where incinerator temperatures averaged 788°C (1450°F), to detect high levels of mercury in the stack emissions. Studies by Whitmore⁵ and Sebastian,^{6,7} however, reported that 50% of the mercury in the feed sludge is retained in the incinerator ash, and that only 10% is emitted to the atmosphere. Olexsey,⁸ on the other hand, did not detect mercury in the ash of three wastewater treatment plants examined, thus suggesting that all of the mercury is either emitted or picked up by the scrubber water. This lack of agreement is not unexpected, because a common basis for comparison does not exist; that is, the literature data were generated only from partial mass balances, where emissions were obtained by "subtraction" as opposed to sampling.

During the three mass balance studies conducted at Northwest Bergen, mercury that

TABLE II. M

Sampling Period	Sluc Fee
Zinc	696
g of total	100
Cum. %	—
Cu	506
g of total	100
Cum. %	—
Pb	150
g of total	100
Cum. %	—
Cr	18
g of total	100
Cum. %	—
Ni	12
g of total	100
Cum. %	—
Cd	2
g of total	100
Cum. %	—

* ND = glass fiber
 b Net metal weight

was unaccounted is felt that this tributary to flow cury detection l water, or emiss with accurately and mercury lev heterogeneous g this heavy meta tity was consid balance calculat rates the mercu operating day.

SUMMARY

Actual total r examined, as sh

TABLE III. M fluidized bed sl

Metal	Sluc Fee
Cd	
Cr	
Cu	11
Pb	3
Ni	
Zn	15

* ND = Glass f

TABLE II. Mass balance—sampling period summary—Northwest Bergen.

Sampling Period	A (7:15-11:15 a.m.)				B (11:15-3:15 p.m.)				C (3:15-5:30 p.m.)			
	Sludge Feed	Ash	Scrubber Water ^b	Stack	Sludge Feed	Ash	Scrubber Water ^b	Stack	Sludge Feed	Ash	Scrubber Water ^b	Stack
Zinc	696	320.6	87	ND ^a	549	264	70.3	ND	346.9	184.4	39.3	ND
% of total	100	46.2	12.5	—	100	48.1	12.8	—	100	53.2	11.3	—
Cum. %	—	46.2	58.7	58.7	—	48.1	60.9	60.9	—	53.2	64.5	64.5
Cu	506	293	85.9	0.5	419	24.3	75.5	0.1	264.4	171.7	40.7	0.1
% of total	100	57.9	16.9	0.1	100	57.4	18.0	0.02	100	64.9	15.4	0.04
Cum. %	—	57.9	74.8	74.9	—	57.4	75.14	75.16	—	64.9	80.3	80.3
Pb	150.2	137.6	20.6	0.4	112.7	95.3	15.8	0.08	76.2	73.7	7.9	0.2
% of total	100	91.6	13.7	0.3	100	89.6	14.0	0.07	100	96.7	10.4	0.3
Cum. %	—	91.6	105.3	105.6	—	89.6	103.6	103.7	—	96.7	107.1	107.4
Cr	18.4	12.2	0.4	0.2	16.3	14.3	0.5	0.1	8.8	7.5	0.3	0.04
% of total	100	66.3	2.1	1.1	100	87.7	3.1	0.6	100	85.2	3.4	0.4
Cum. %	—	66.3	68.1	69.2	—	87.7	90.8	91.4	—	85.2	88.6	89.0
Ni	12.3	8.0	0.7	0.03	11.6	5.4	0	0.03	6.3	3.8	0.05	ND
% of total	100	65.0	5.7	0.2	100	46.6	0	0.3	100	60.3	0.8	—
Cum. %	—	65.0	70.7	70.9	—	46.6	46.6	46.9	—	60.3	61.1	61.1
Cd	2.1	1.5	0.4	0.002	1.3	1.2	0.06	ND	1.1	0.8	0.4	ND
% of total	100	71.4	19.0	0.1	100	92.3	4.6	—	100	72.7	36.4	—
Cum. %	—	71.4	90.4	90.5	—	92.3	96.9	96.9	—	72.7	99.1	99.1

^a ND = glass fiber filter background exceeded sample amount.
^b Net metal weight (grams): scrubber water out (sampling point 4) - scrubber water in (sampling point 2).

was unaccounted for averaged about 20%. It is felt that this closure problem was not attributable to flow measurement errors or mercury detection limitations in the ash, scrubber water, or emissions. Rather, it is associated with accurately measuring feed sludge volumes, and mercury levels in the thickened sludge (a heterogeneous gelatinous material). Thus for this heavy metal only, the air emission quantity was considered as the "total value" for balance calculation purposes. Figure 2 illustrates the mercury balance for the incinerator operating day.

SUMMARY

Actual total recoveries for the heavy metals examined, as shown in Table III, ranged from

60 to 104%. Considering that these mass balances were conducted during a normal plant operating day, as opposed to controlled conditions in a laboratory or pilot plant, the recovery values reported are acceptable. Table IV summarizes these data, based on a normalization to 100% of the actual field recoveries.

It is important to note that, although most of the mercury contained in the feed sludge was emitted to the atmosphere during this test program, the Northwest Bergen facility did not violate the EPA mercury emission limit of 3 200 grams per day. Other wastewater sludge incinerators are likely to violate this standard if higher levels of mercury are contained in the raw wastes.

TABLE III. Mass balance total "day" operation at Northwest Bergen's fluidized bed sludge incinerator.

Metal	Sludge Feed (g/d)	Ash (g/d)	Scrubber (Net) (g/d)	Stack (g/d)	Total % Recovery	% Wgt. Remain Ash	% Wgt. Scrubber	% Wgt. Atmos.
Cd	4.4	3.5	0.9	ND ^a	100	79.5	20.5	ND ^a
Cr	43	34	1.2	0.3	82.6	79.1	2.8	0.7
Cu	1 189	705	203	1.0	76.5	59.3	17.1	0.1
Pb	339	307	45	0.7	104.0	90.5	13.3	0.2
Ni	30	17	0.8	0.03	59.5	56.7	2.7	0.1
Zn	1 592	770	197	0	60.8	48.4	12.4	0.0

ND = Glass fiber filter background exceeded sample amount.

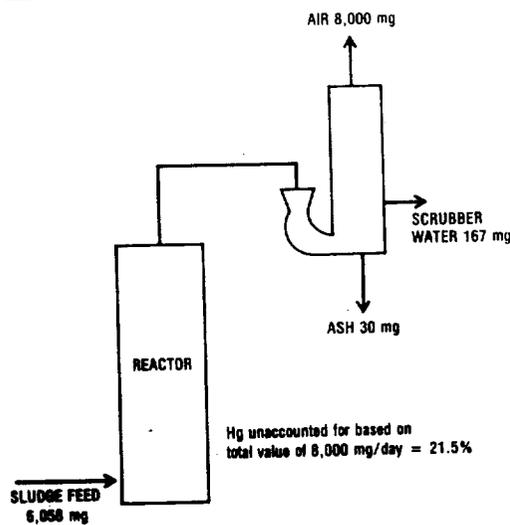


FIGURE 2. Mercury mass balance for operating day (10.25 hours) Northwest Bergen.

With the recent promulgation by EPA of a national ambient standard for lead— $1.5 \mu\text{g}/\text{m}^3$ —and the future possibility of a cadmium standard, additional studies are needed to substantiate the possible loss of these metals to the atmosphere. Laboratory and pilot studies in pyrolysis-type incinerators by Mytelka⁹ and Takeda¹⁰ suggest significant losses—30–50% of both metals, particularly when temperatures exceed 649°C (1200°F). However, studies by Copeland¹¹ support the findings of this investigation; that is, no significant loss of either cadmium or lead. A possible explanation for these reported differences would appear to be centered in the sampling train used for measuring stack emissions. Method 5, unless modified (as done during this study for only mercury), is not

TABLE IV. Fluidized bed incinerator—heavy metal mass balance, % weight distribution (normalized).

Metal	Ash	Scrubber	Stack
Zinc	79	20	1
Copper	78	21	1
Lead	87	12	1
Chromium	95	4	1
Nickel	80	20	ND*
Mercury	0.4	2	97.6
Cadmium	80	20	ND*

* ND = Glass fiber filter background exceeded sample amount.

designed to capture submicron particles that may pass through the glass fiber filter.

CONCLUSIONS

• With the exception of mercury, there is no evidence to suggest significant Cd, Cr, Cu, Ni, Pb and Zn enrichment of the off-gas and particulates emitted from a fluidized bed wastewater sludge incinerator. Ash contained, depending upon the metal, from 78 to 95% of the total amount present in the feed sludge. Scrubber water from the incinerator contained from 4 to 21% of the particular heavy metal.

• Approximately 98% of the mercury in the feed sludge was emitted to the atmosphere when combustion temperatures averaged 788°C (1450°F). Only 0.4% was retained in the ash and slightly more than 2% was found in the scrubber water.

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