

Accepted for publication in Environmental Technology (1990) |CHANGES AND VARIABILITY IN CONCENTRATION
OF HEAVY METALS IN SEWAGE SLUDGE DURING COMPOSTINGD.J. Wagner, G.D. Bacon, W.R. Knocke¹, and M.S. Switzenbaum*Department of Civil Engineering, University of Massachusetts
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ABSTRACT

The heavy metal concentration of sewage sludge increased during composting for cadmium, chromium, lead and zinc and decreased for copper and nickel. Cadmium and lead were the most variable of the metals, copper and zinc the least. Variability within a batch was small (variation of 4-12%). Variability attributable to batch-to-batch differences was generally larger (variation of 4-37%). The variability of sludge entering the batch process and of compost exiting the process were similar for each metal.

INTRODUCTION

Environmental problems associated with sewage sludge disposal have prompted strict legislative actions over the past 20 years. At the same time, upgrading and expansion of wastewater treatment plants has greatly increased the volume of sludge generated. As a consequence of stricter regulations, greater sludge production, and decreasing availability of landfill space, new practices for sludge treatment and resource recovery from sludge have been encouraged. Sludge composting is among the most promising of these practices (1). Sewage sludge composting can provide: 1) organic matter stabilization, 2) pathogen destruction, and 3) mass, volume and moisture content reduction (2,3). Composted sludge is aesthetically acceptable, essentially pathogen free, easy to handle, and valuable as a soil amendment.

A significant obstacle to the beneficial land application of sludge or compost is heavy metal contamination (4-6). Metals are removed from wastewater during primary and secondary treatment by adsorption onto organic particles and to microbial surfaces (7,8). These organic-metal particles as well as inorganic metal precipitates become incorporated into sludge. The type of land feasible for sludge disposal (private home property, agricultural land, non-agricultural land) and the sludge loading rate permissible become more restrictive as the metal concentration of sludge or compost increases.

Composting can concentrate or dilute the heavy metals present in sewage sludge. The change in metal concentration should depend on: 1) incorporation of organic bulking agent into the final compost, 2) destruction of sludge organic matter by thermophilic microorganisms, 3) addition of metals from

contaminated bulking agents, 4) metal loss through leaching, and 5) physical/chemical interactions between compost and bulking agent. The few reports available in the literature on changes in heavy metals concentrations during composting show both dilution and concentration depending on the bulking agent (9-12). The sampling regimes used in these studies were generally not designed to measure the variability of sludge and compost or to make rigorous statistical tests on concentration changes through the composting process. Information on concentration changes and variability is fundamentally important in selecting a sludge disposal option and in specifying compliance monitoring sampling programs.

Objectives and Approach

This study had two primary objectives: 1) to determine if significant changes in the heavy metal concentration of sludge occur during composting, and 2) to measure the variability of sludge and compost, a) within a given batch-processed pile, and b) from batch to batch over several months.

These objectives were addressed by establishing a sludge and compost monitoring and sampling program for a well controlled, sequential, batch composting process at a single wastewater treatment plant. Six metals, cadmium, chromium, copper, nickel, lead and zinc, were monitored. Thirteen compost batches were sampled over six months to evaluate variability and concentration changes. The experimental design allowed rigorous statistical analysis of any changes in metal concentrations detected.

The sampling program in this study was aimed strictly at monitoring concentrations of metals in sludge and compost. No attempt was made to perform a mass balance on metals or solids.

MATERIALS AND METHODS

Process Description

The Hoosac Water Quality District treatment plant (Williamstown, MA) was selected for this study for three reasons: 1) a well established aerated static pile composting process (operating since 1979), 2) a well defined, sequential, batch process amenable to monitoring parent sludge entering a pile and final compost produced, and 3) a history of metal contamination of sludge. The plant employs conventional activated sludge treatment with co-settling of primary and secondary sludge in the primary clarifier. The average daily flow is $21,200 \text{ m}^3 \text{ day}^{-1}$ and sludge production is about $50 \text{ m}^3 \text{ day}^{-1}$.

Prior to composting, sludge is lime treated and dewatered on vacuum filters to 15-18% solids. The dewatered sludge is conveyed directly to a truck where it is mixed with wood chips for bulking (wood chip to sludge ratio 2:1 by volume). The truck deposits the sludge-wood chip mixture into one of eight open ended concrete bins (21.4 m x 4.5 m x 3.7 m). Each bin takes 2 to 4 days to fill. Compost is aerated by mechanical blowers connected to grate covered channels running the length of the bin. For the first 6-8 days of composting, compression and selfheating in the pile released leachate. The leachate is carried away by the aeration channels. Composting time is typically 33 ± 3 days. Each filled bin is designated as a batch.

When composting is complete, a front end loader empties the bins via the open end. From June to November, the mixture is immediately separated by mechanical screening through 1.25 cm mesh. Compost is stockpiled for curing; wood chips are recycled. From December to May, compost does not lose enough water to allow efficient screening. The mixture is therefore stockpiled for later screening. During these cold months new wood chips are used for each new pile.

Sample Collection

Two types of samples, filter cake from the vacuum filters (sludge samples) and wood chip-compost mixture from the composting bins (compost samples), were collected during this study (6). The sampling regimes were designed to accurately represent the material entering and leaving a given batch. Between 25 January 1988 and 24 May 1988 every other bin filled at the plant was sampled (total of 13 batches). Sludge samples were collected in 120 mL polyethylene containers directly from the vacuum filters at two hour intervals. For most batches, a maximum of four samples were collected per day for the 2 to 4 days required to fill a bin. The total number of influent sludge samples collected per compost batch ranged from 11 to 20.

Compost samples were collected at two sections as the bin was emptied, 50% empty and 90% empty. At each section, three or four samples (packed in 500 ml polyethylene containers) were collected on a transection from the bottom to the top of the exposed compost pile face. Since bins were filled evenly over their entire length, the bottom to top transection reflected any temporal variation in the sludge added to the bin. Both sludge and compost samples were collected by plant personnel. Samples were refrigerated at 4°C until processed.

Sample Processing and Analysis

Sample preparation consisted of drying, grinding and acid digestion. Sludge was prepared by drying an aliquot of each sample at 103°C, grinding the sample with a mortar and pestle and redrying the fine power at 103°C. For composted samples, sieving was chosen as the method to separate wood chips and wood fragments from the composted sludge particles. The entire 500 mL of each compost sample was first dried at 103°C for 48 hours. Next the sample was placed on a 2 mm sieve with ten 1.25 cm porcelain balls and shaken for 15 minutes. The balls provided agitation to help dislodge dry sludge from the wood chips. Material passing the mesh was sieved gently twice more by hand without the balls to remove any remaining splinters of wood. Sieved compost was ground with a mortar and pestle and redried at 103°C.

Screened compost prepared by the mechanical screens at the plant was tested with the laboratory sieving method. About 95% of each sample passed the 2 mm sieve. This indicated that the laboratory and plant processes produced similar size differentiation.

Aliquots of dried samples (0.50 g) were digested with nitric acid according to the protocol of Thompson and Wagstaff (13). Digested samples were brought to the 50 mL volume (ca. 10% acid by volume) and filtered through Whatman GF/C filters (1.2 micron effective retention). Metals concentrations (Cd, Cr, Cu, Ni, Pb, and Zn) were measured by flame atomic adsorption spectrophotometry with a Perkin-Elmer Model 3030B according to the manufacturer's recommendations. A single digested aliquote was analyzed for each sample. Sludge metals concentrations in mg metal per kg dry weight were calculated from digest concentrations. These data were analyzed by standard analysis of variance techniques (14).

RESULTS AND DISCUSSION

Changes During Composting

Heavy metals concentrations changed during sewage sludge composting (Fig. 1, Tables 1 and 2). The concentrations of Cd, Cr, Pb and Zn were higher in compost than in parent sludge while the concentrations of Cu and Ni were lower. The degree of change was different for each metal. Cadmium concentrations changed most during composting and copper concentrations least (Table 1). For all metals, these changes were highly statistically significant (Table 2, sludge type column). The amount of concentration or dilution of each metal varied by batch. This phenomenon was also statistically significant as indicated by the batch X sludge type interaction (Table 2).

Table 1. Percent change in metals concentrations during composting, range and average over all batches.

Metal	Average Change (%)	Range of Change (%)
Cd	58	9 to 100
Cr	21	7 to 47
Cu	-8	-13 to -2
Ni	-11	-19 to 4
Pb	16	-8 to 43
Zn	12	0 to 24

Table 2. Statistical significance⁺ of changes in metals concentrations during composting.

Metal	Sludge Type	Batch	Batch x Sludge Type Interaction
Cd	***	***	***
Cr	***	***	***
Cu	***	***	*
Ni	***	***	***
Pb	***	***	***
Zn	***	***	*

* significant F statistic, $p < 0.05$

*** very highly significant F statistic, $p < 0.001$

⁺A separate analysis of variance was carried out for each metal. The analysis matrix was: 2 sludge types x 11 batches with 6 or 8 values in each compost cell and 11 to 20 values in each sludge cell. In all tests a pooled, unweighted, estimate of within cell variance was used (14).

The systematic increase in concentration of certain metals during composting while other metals decreased in concentration indicated that the changes were not due solely to the gain or loss of sludge solids. Simple concentration from sludge organic matter destruction or simple dilution from incorporation of bulking agent into compost should change the concentration of all metals by the same percentage. Removal of metals sorbed onto bulking agents and metal removal in leachate from the piles are possible causes for the differential changes in metal concentrations. Metal concentrations in the new woodchips bulking agent used during this study were too low (below analysis method detection limits) to affect metal concentrations in the final compost.

A reasonable scheme that accounts for the observed concentration changes during composting is: 1) overall concentration of metals due to organic matter destruction and good separation of compost and bulking agent and 2) greater leaching of certain metals. Cheng *et al.* (15) and Sterritt and Lester (16) reported the affinity of metals for sludge is $Cr > Cd > Ag > Pb > Zn > Cu > Ni$. This

supports a leaching hypothesis for our composting study since the metals with greatest affinity had the highest percent increase in concentration. The metals with the lowest affinity, Cu and Ni, showed apparent dilution (Table 1).

Influence of Bulking Material

The potential contribution of metal adsorption onto wood chips to the differential changes in metal concentrations during composting is difficult to evaluate. Benson (17) showed that heavy metals from a pH 4 simulated landfill leachate solution adsorbed differently to sawdust during a column testing procedure (Cr>Cu>Zn>Pb>Cd). The pH of leachate from the compost piles was approximately 11.5 due to lime addition. The order of metal affinity to woodchips is probably different at this high pH. Heavy metals probably adsorb differentially to wood chips under composting conditions but it is not possible to judge how much of the observed metals concentration changes were due to adsorption.

Other studies of heavy metal concentration changes during composting with wood chips, generally showed lower metals concentrations in compost than in sewage sludge (9-12). This contrasts with our study where four of the six metals monitored increased in concentration during composting. The metals that concentrated the most during this study, Cd (58%) and Cr (21%), showed strong dilution (-40% to -89%) in pilot scale composting tests with wood chips, brush chips and bark as bulking agents (10,11). The weakly concentrated metals in our study, Pb (16%) and Zn (12%) also showed strong dilution (-40% to -79%) in the pilot studies. A likely reason for dilution in the pilot studies was greater incorporation of bulking material into the final compost than occurred during our study.

As in this study, metal concentrations changed differentially during pilot scale composting of sewage sludge (9-12). However, for any given pilot study percent changes were generally more similar among the metals monitored (Cd, Cr, Cu, Ni, Pb, Zn) than in our study. For example, in a wood chip/sewage sludge composting project (9) the difference between the most diluted metal (Cu at -66%) and the least diluted metal (Zn at -43%) was only 23%. In contrast, the difference was 69% in our study, from Cd at +58% to Ni at -11% (Table 1). It seems that greater dilution from incorporation of bulking material and less differential leaching from the piles was occurring in the pilot composting projects.

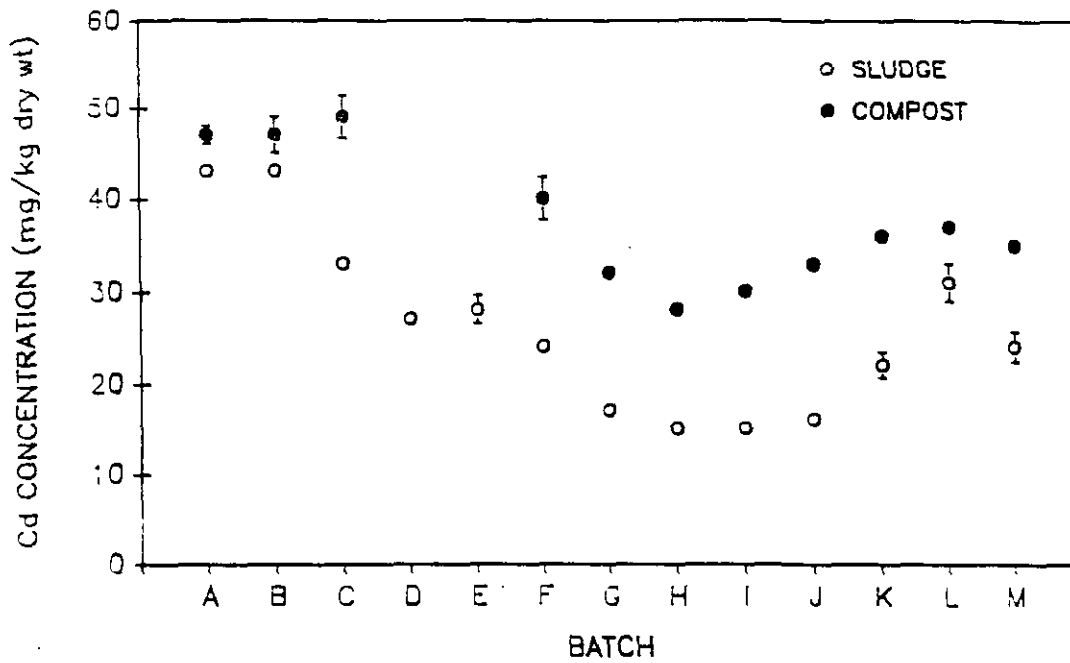
Changes Among Batches

Changes in sludge and compost metal concentrations occurred over the five month sampling period (Fig. 1). Changes in cadmium concentration probably reflected changing inputs from a film manufacturer that discharged into the sewer system. A relatively constant input of copper from household plumbing may be responsible for the small seasonal changes in sludge and compost copper levels. There are no obvious explanations for the temporal changes of other metals. Between batch change was highly statistically significant for all metals (Table 2, batch column).

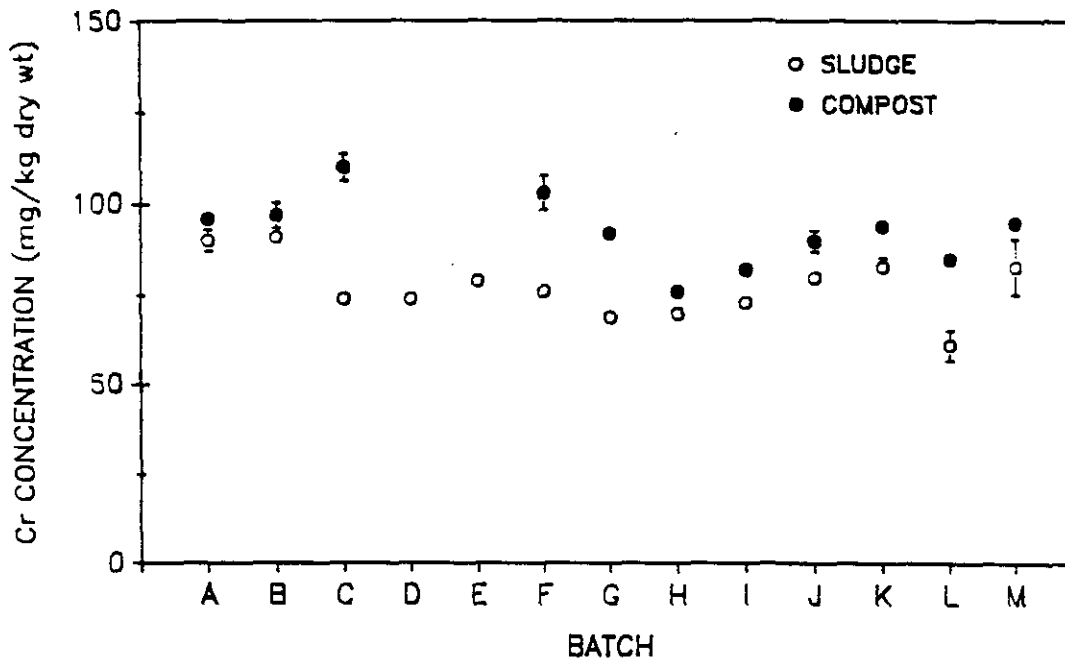
Variability of Sludge and Compost

Within a batch, metal concentration variability in sludge and compost ranged from 4 to 12% as measured by the coefficient of variation (Table 3). The within batch standard deviations and coefficients of variation represent the metals variability over a ca. 3 day period. The within-batch variability of sludge and compost were quite close for each metal tested. This suggests that the daily variation in sludge metal concentrations was preserved in the vertical profile during composting. Variation among batches was generally larger than variation within batches (CV range from 4 to 37%). The among-batch standard deviations and coefficients of variation represent metal variability over the

Figure 1. Changes in heavy metal concentrations in sludge and compost.



AVERAGE (\pm STD ERROR) CADMIUM CONCENTRATIONS IN SLUDGE AND BATCH COMPOST SAMPLES



AVERAGE (\pm STD ERROR) CHROMIUM CONCENTRATIONS IN SLUDGE AND BATCH COMPOST SAMPLES

Figure 1. continued

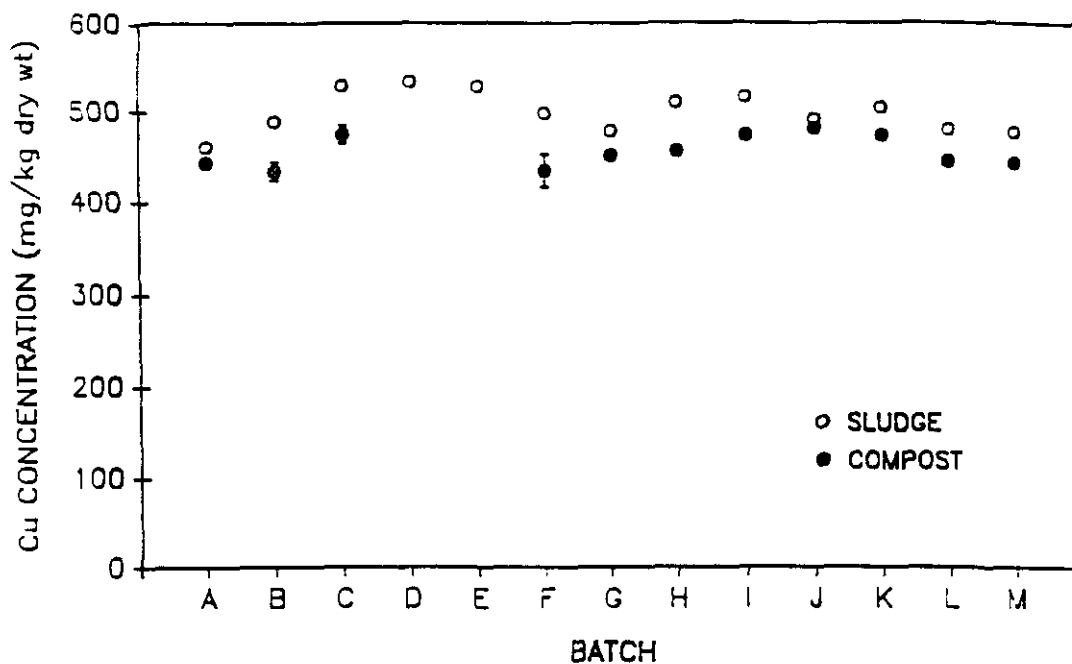
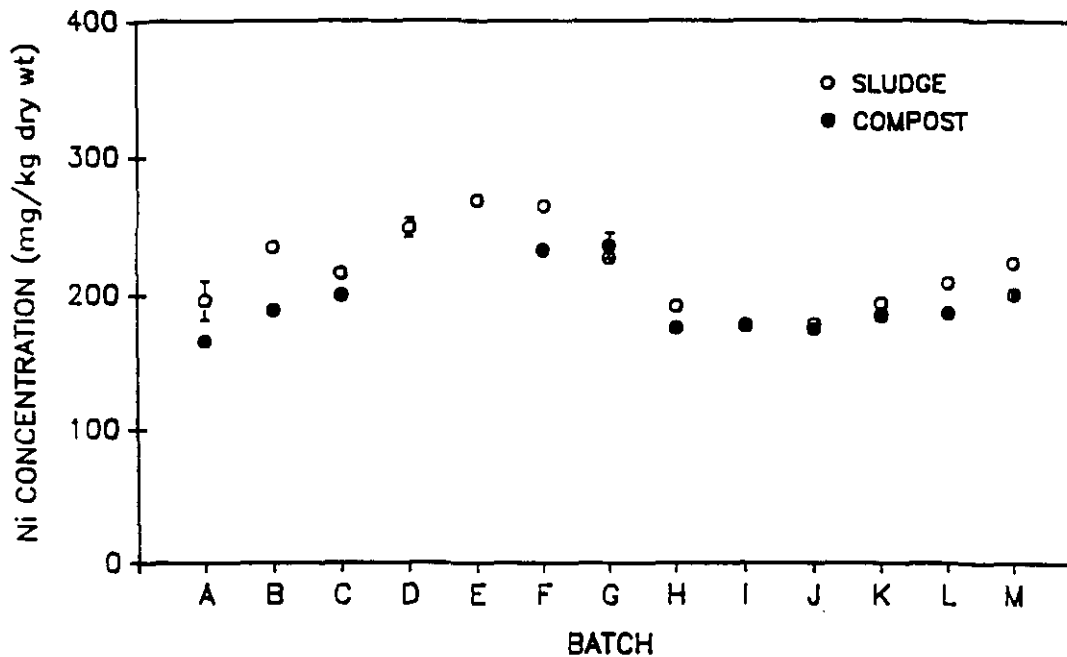
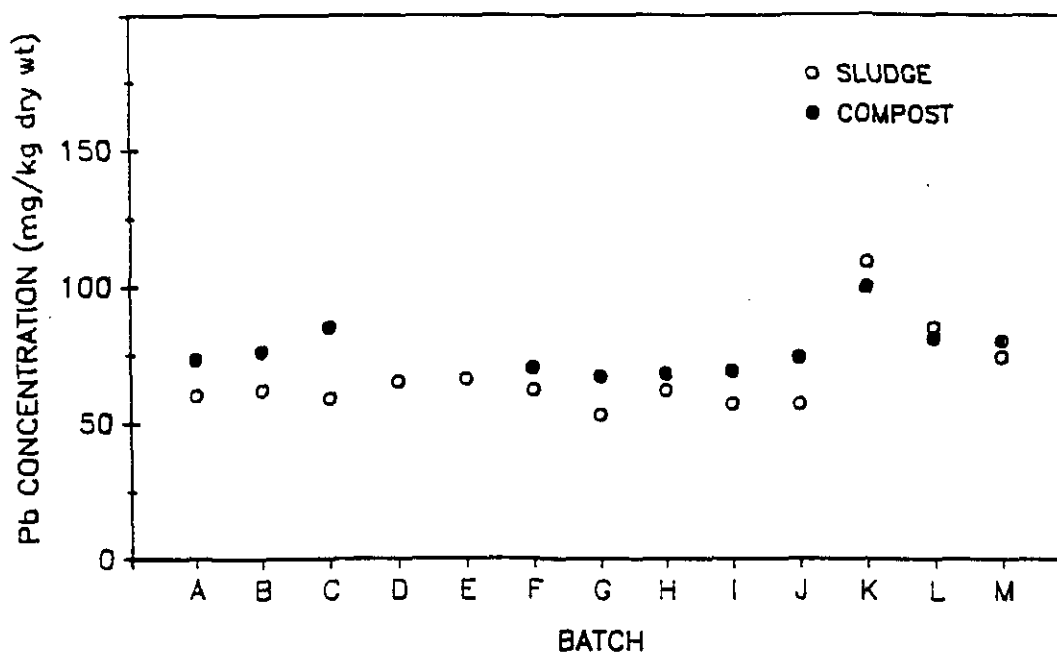
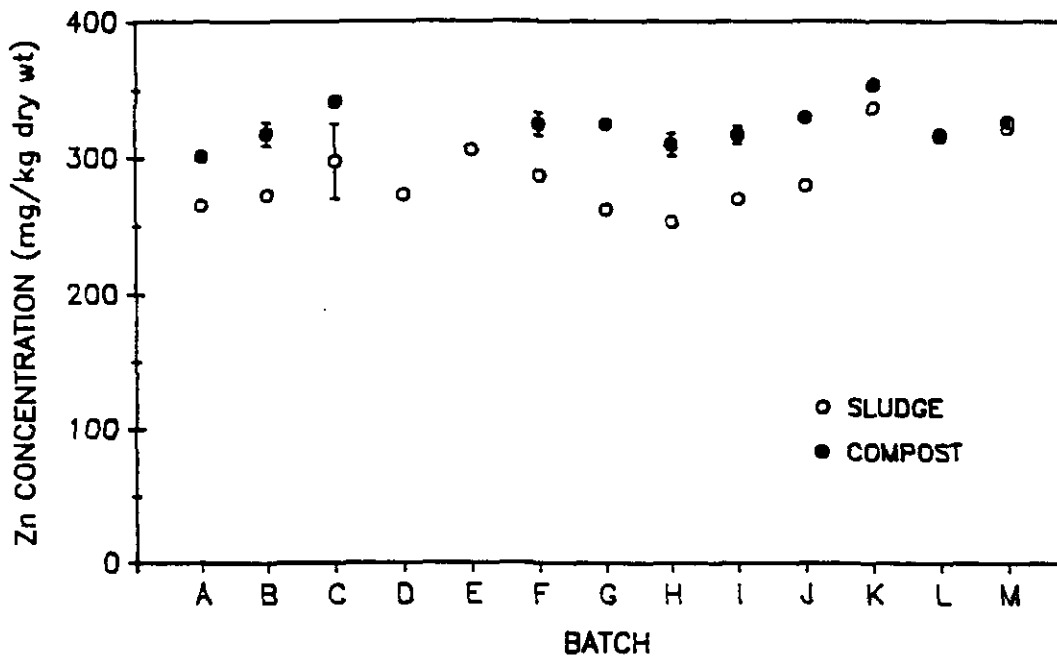
AVERAGE (\pm STD ERROR) COPPER CONCENTRATIONS
IN SLUDGE AND BATCH COMPOST SAMPLESAVERAGE (\pm STD ERROR) NICKEL CONCENTRATIONS
IN SLUDGE AND BATCH COMPOST SAMPLES

Figure 1. continued



AVERAGE (\pm STD ERROR) LEAD CONCENTRATIONS IN
SLUDGE AND BATCH COMPOST SAMPLES



AVERAGE (\pm STD ERROR) ZINC CONCENTRATIONS IN
SLUDGE AND BATCH COMPOST SAMPLES

Table 3. Variation in metals concentrations within and between batches. SD is Standard Deviation. CV is Coefficient of Variation.

Variation Within Batches *						
Metal	SLUDGE			COMPOST		
	Mean (mg/kg)	SD (mg/kg)	CV (%)	Mean (mg/kg)	SD (mg/kg)	CV (%)
Cd	26	3.7	12	38	3.6	8
Cr	77	9.5	10	93	6.8	6
Cu	500	22.0	4	456	22.0	4
Ni	218	16.5	6	193	10.8	4
Pb	67	7.2	10	77	5.5	6
Zn	288	14.4	5	324	16.4	5

Variation Between Batches **				
Metal	SLUDGE		COMPOST	
	SD (mg/kg)	CV (%)	SD (mg/kg)	CV (%)
Cd	9.6	37	7.2	19
Cr	8.4	11	9.5	11
Cu	23.1	5	18.3	4
Ni	30.6	14	22.9	12
Pb	15.1	22	9.7	13
Zn	25.9	9	14.6	4

*The SD is the square root of pooled bin variance. CV is each batches' standard deviation/mean averaged across batches.

**Values calculated from bin means.

five month study period. The between-batch coefficients of variation are smaller than those measured by Sommers (18), 32 - 72% CV, and Doty *et al.* (19), 21 - 47% CV, for sludges sampled from one wastewater treatment plant over many months.

The pattern of variability of a few of the metals is noteworthy: 1) Cadmium and lead had the highest variability both within and between batches. 2) Copper and zinc had the lowest within and between batch variability. 3) For copper and zinc the within batch variability was about the same as the between batch variability.

The relatively low within-batch variability suggests that a modest sampling plan could be used to characterize the metals concentrations in a particular batch. For this plant, 3-5 samples of compost taken from a vertical transection of a pile or a single sample composited from a vertical transection should adequately represent a compost batch. Characterizing the sludge and compost quality seasonally must be based on the most variable metals concentrations. Based on cadmium and lead, sampling every fourth batch (about every 15 days) would appear adequate. Because of the concentration changes that take place during composting, samples of the sieved compost must be collected for analysis. Monitoring all the potential metal contaminants during composting is important, because the dilution or concentration factor for each metal is different.

IMPLICATIONS

The results of this study have two practical implications:

1) The increase in metals concentrations detected during composting implies that a parent sludge that meets requirement before composting may not meet requirements after composting. This is especially true for cadmium which showed an average increase in concentration of 58% in this study. Cadmium is often the critical metal in determining the sludge/compost disposal or distribution options. Some control of the degree of metal concentration might be possible by making amendments to the bulking agent. For example, addition of a readily degradable or easily fragmented amendment to ordinary wood chips (eg. leaves, brush chips, sawdust) would effectively decrease the final compost metal concentration. Leaving the bulking agent mixed with the final compost would accomplish the same thing.

2) The significant variation in sludge and compost heavy metal concentrations implies that regulations and permits should specify sampling regimes with adequate sampling frequency and specific sampling methods. For example, the treatment plant sampled during this study was required to take quarterly samples of compost. This sample was composited from the surface of a curing stockpile which consisted of four to six months of accumulated compost. This type of sample fails to reflect current compost quality and fails to gauge seasonal changes in compost quality. Based on our study, a sampling program with the following characteristics would be sound: 1) sample frequency twice per month, 2) random designation of certain piles or compost batches at the beginning of each month to be sampled as the finished compost is screened, and 3) compositing the compost sample by subsampling across the depth and length of the pile or batch. This type of sampling regime provides sound monitoring of current sludge quality and detects any seasonal changes in sludge quality.

CONCLUSIONS

Several important conclusions can be formulated from this study:

.Concentrations of cadmium, chromium, lead and zinc in sludge increased (12% to 58%) during composting with wood chips, while concentrations of copper and nickel in the final compost decreased (8% to 11%). The data suggest that a combination of metal concentration from sludge solids destruction and

differential leaching of metals from the pile could account for the observed changes.

Heavy metals concentrations fluctuated over the five month course of the study. Temporal variation ranged from a high of 37% for sludge cadmium concentration to a low of 5% for copper concentrations.

Some characteristics of sludge and compost heavy metal variability became evident:

- Within-batch variability was relatively small (4% to 12%) for all metals.
- For each metal, within-batch variability of sludge and compost were similar.
- With the exception of copper and zinc, between batch variability was greater than within-batch variability.
- The most variable metals concentrations were cadmium and lead. The least variable were copper and zinc.

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