Nature Environment and Pollution Technology
An International Quarterly Scientific Journal

p-ISSN: 0972-6268
e-ISSN: 2395-3454

Vol. 17 | No. 3 | pp. 999-1004 | 2018

Original Research Paper | Open Access

Fractionation and Reduction in Bioavailability of Toxic Heavy Metals During Rotary Drum Composting of Paper Mill Sludge

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Nat. Env. & Poll. Tech.
Website: www.neptjournal.com

Received: 08-12-2017
Accepted: 17-01-2018

Key Words:
Paper mill sludge
Rotary drum composting
Heavy metals
Chemical speciation
Bioavailable factor

ABSTRACT

Composting is the most suitable recycling option for paper mill sludge (PMS) due to high organic content. However, final compost produced from PMS contain high concentration of heavy metals, resulting in restricted land application of compost. The aim of the study was to reduce bioavailable form of heavy metals during the composting of PMS using rotary drum composter. Therefore, this study was carried out to identify the variation in distribution of different fractions (exchangeable, carbonate, reducible, oxidizable and residual fraction) of Cd, Pb and Cr during rotary drum composting of PMS. Tessier sequential extraction method was applied for speciation of heavy metals. Residual fraction of most of heavy metals was found to be dominant fraction in all treatments. The concentration of Cd was not very high; however, its bioavailable fractions were found to be high. This study shows that addition of cattle manure in appropriate quantity can influence the physico-chemical parameters, resulting in decrease in the bioavailable fraction (exchangeable and carbonate) of heavy metals during composting process. An addition of cattle manure decreased the exchangeable and carbonate fractions of metals during the composting process. A reduction in bioavailability factor was also observed in all treatments during the composting process.

INTRODUCTION

The use of paper by a society is often taken as an index of its development. The rapid industrialization and development is accountable for the increase in paper consumption. By 2025, the projected global consumption of paper will be 500 million tonnes, with existing production of 394 million tons per annum (Kujur 2012), which is expected to increase by 77% by the year 2020 (Likon & Tresbe 2012). Approximately a paper mill generates 40-50 kg of dry sludge during the production of paper per tonne (Bajpai 2015).

An increase in paper industries will result in generation of high amount of sludge, making sludge management, a serious concern. Currently about 69% of total sludge produced goes for land filling, whereas 21% goes for incineration, 8% for land spreading, and 8% for other alternatives (Scott & Smith 1995). Composting is the most suitable recycling option for PMS due to the high organic content of the sludge. Composting also removes the short term phytotoxic effects (Line 1995), increases cation exchange capacity and nutrient retention efficiency of soil (Sesay et al. 1997), and kills the pathogenic bacteria present in it.

The elevated heavy metal concentration of PMS is a serious concern as it can cause serious environmental problems after land application (Singh & Kalamdhad 2013a). A very small amount of heavy metals is consumed by the plants for metabolism, but concentration exceeding the optimum limit becomes toxic to plants. Heavy metals present in soil can affect the metabolism, which in turn hinders the growth and morphology of soil microbes and consequently loss of soil fertility (Bragato et al. 1998). Total concentration of heavy metals present in the final compost is a very reliable overall pollution indicator, but it cannot provide information about bioavailability of the metals.

Chemical speciation is the process for identifying and quantifying different species, forms or phases present in a composting material (Cai et al. 2007). Chemical speciation is a very reliable technique for determining the chemical forms in which the metals are present (Walter et al. 2006, Singh & Kalamdhad 2012). The objective of this work is to determine the total concentration of heavy metals (Cd, Pb and Cr), evaluate the contribution and variation of different fractions through sequential extraction procedure and assess the effect of physico-chemical parameters on metal speciation during composting of paper mill sludge.

MATERIAL AND METHODS

Raw materials collection: Primary paper mill sludge (PMS), cattle manure and sawdust were mixed in five different ratios for preparing the feed material for rotary drum composting. PMS was collected from Nagaon Paper Mill, a unit of Hindustan Paper Corporation situated at Nagaon in the State of Assam, India at a distance of around 70 km from Indian Institute of Technology, Guwahati (IITG) campus. For treating the wastewater, the mill is using a combination of clariflocculator and aerated lagoon for primary treatment and secondary treatment respectively. The sludge produced from clariflocculator is dewatered using filter press and disposed. The dewatered sludge from filter press was collected for making compost. The dairy farms near the IITG campus supplied the cattle. Sawdust was bought from the saw mills near IITG campus. The bamboo chips were segregated and lumps of the PMS were broken into 1 cm particle size to facilitate better aeration. The proportions and initial characterization of raw materials is given in Table 1.

For making compost out of PMS, a rotary drum composter (capacity 500 L) was used as described by Singh & Kalamdhad (2013b). Proper degradation and stabilization of PMS was the key parameter in fixing the duration of 20 days of composting process. Grab sampling was applied at various positions inside the drum, mainly top, bottom and middle layer for collecting samples. All the sampling was accomplished with the help of a sampler without disturbing the adjacent material. Collected samples were then mixed to get a homogenized sample. Triplicate samples were collected at each 4 day interval till the end of composting process. These samples were dried at 105°C for 24 h and then milled and sieved through 0.2 mm sieve.

Physico-chemical analysis and metal fractionation: Water extract of compost was used to measure pH. Moisture content was determined by heating the sample at 105°C for 24 h. Sequential extraction of the heavy metals in the compost samples has been applied according to the method of Tessier et al. (1979). Five fractions were obtained: (i) exchangeable fraction (F1), (ii) carbonate fraction (F2), (iii) reducible fraction (F3), (iv) organically bound fraction (F4)

Table 1: Mixing proportions of the composting materials.

Parameters	PMS	Cow dung	Saw Dust
Trial 1 (T1) (kg)	150	0	0
Trial 2 (T2) (kg)	120	15	15
Trial 3 (T3) (kg)	105	30	15
Trial 4 (T4) (kg)	90	45	15
Trial 5 (T5) (kg)	75	60	15

and (v) residual fraction (F5). The sequential extraction was performed. The details of extraction method followed as given by Singh & Kalamdhad (2012).

According to the extraction procedure, total concentration of F1, F2, F3 and F4 represents the concentration of mobile fraction (Liu et al. 2007). A ratio (bioavailable fraction) was calculated from the ratio of mobile concentration to the total concentration, to assess the bioavailability of heavy metals.

$$\text{Bioavailable Fraction (BF)} = \frac{F1 + F2 + F3 + F4}{F1 + F2 + F3 + F4 + F5}$$

RESULTS AND DISCUSSION

Physico-Chemical Parameters

The temperature profile of the rotary drum composting of PMS is shown in Fig. 1a. Trials with high amount of cattle manure (trials 4 and 5) are showing a rapid increase in temperature due to high microbial activity, whereas trials 1-3 with comparatively less cattle manure are attaining the maximum temperature at later stage during composting. Maximum temperature of 44.8°C was observed in trial 4. In trial 1, increase in temperature was not observed due to absence or low microbial activity. It is believed that a temperature between 52°C and 60°C is enough to continue thermophilic activity (Mohee & Mudhoo 2005). Varma et al. (2015) reported a maximum temperature of 61.4°C in rotary drum composting of vegetable waste. Thus, the maximum temperature in PMS composting depicts that the material is hard to degrade. The initial sharp increase in the temperature is due to high microbial activity which may be caused due to the addition of cattle manure. The prolonged consistent temperature from middle to final days depicts that the effect of the cattle manure is minimized, but biodegradable organic was still present in the sludge which is being used by the microorganisms at a slower rate.

In composting, one of the important environmental factors is the pH. The optimum pH for the sustainability and the degradation efficiency of the microorganisms is in the range of 7-8 (Nakasaki et al. 1993). The pH profile during composting is shown in Fig. 1b. A maximum increase in pH from 7.6 to 8.1 was observed in trial 4 during the composting process, whereas, in trial 1 change in pH was not observed. During the composting process, regular turning was applied to provide sufficient aeration to decrease CO_2 level in the compost resulting in increase in pH, which prevented the development of anaerobic condition (Haug 1993).

Variation in moisture content during composting is shown in Fig. 1c. Moisture loss was observed in all the trials, with a maximum of 23.2% in trial 4, whereas minimum

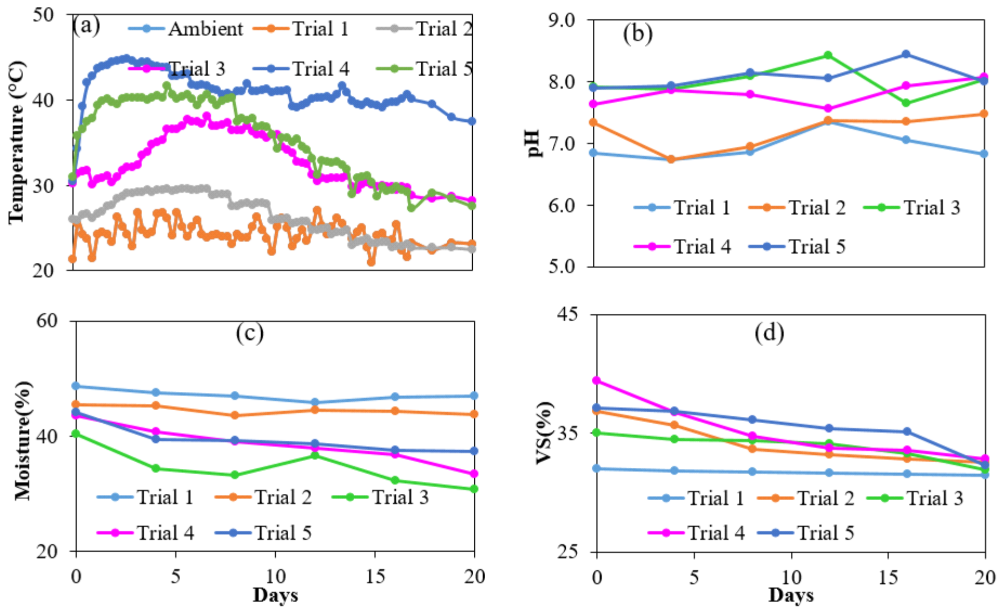

Fig. 1: Variation in physico-chemical parameters during rotary drum composting: temperature (a), pH (b), moisture content (c) and volatile solid, VS (d).

3.4% was observed in trial 1, representing maximum organic matter degradation in trial 4. Decomposition during composting can be assessed by the moisture loss due to the heat generated during the process (Kalamdhad et al. 2008).

During composting, organic matter (OM) is degraded and converted to humic substances. Metal ions react with the humic substances, which can buffer the pH and will serve as a source of nutrients for plants (Amir et al. 2005). The scenario of reduction of OM is presented in Fig. 1d. OM was reduced in all the trials. In trial 4, a maximum reduction of 16.8% was observed, while minimum reduction of 1.9% of OM was observed in trial 1. Higher OM loss in trial 4 was due to higher microbial activity during composting. Singh & Kalamdhad (2013a) has reported a decrease of 32.6% in OM during rotary drum composting of water hyacinth. A less OM reduction in PMS composting can be attributed to the presence of lignocellulosic matters.

Speciation of Heavy Metals

Cadmium: The variation in distribution of different fractions of Cd during composting is shown in Fig. 2. The exchangeable (F1) fraction was found to increase in trial 1, 2 and 4 from 17.5, 5.8 and 10.6 to 18.8, 7.8 and 13.9% respectively. The F1 fraction was not observed in trial 3, whereas in trial 1 it was decreased from 6.3 to 5.2%. Liu et al. (2007) and Singh & Kalamdhad (2012) also observed an increase in F1 fraction during the composting process. The carbonate (F2) fraction slightly decreased in trial 1, 3 and 4, whereas it marginally increased in trial 2 and 5. Liu et al. (2007) have reported an increase in carbonate fraction during composting of sewage sludge. The reducible (F3) fraction remained almost constant in all the trials during composting period. In trial 3, reducible fraction goes below detection limit after the composting period. Oxidizable (F4) fraction was not present in trial 4, while it remained unchanged in trial 1 and 2. In trial 3 and 5, it gradually decreased till the end of the composting process. The residual (F5) fraction was the predominant concentration in all the trials. Residual concentration increased in trial 1 and 3, but decreased in trial 2, 4 and 5. A decrease in residual fraction was reported by Liu et al. (2007) and Singh & Kalamdhad (2012). The increase in residual fraction can be attributed to the ability of Cd to form strong chemical bonds with organic matter (Singh & Kalamdhad 2012).

Lead: The distribution of different species during PMS composting is shown in Fig. 3. F5 fraction was the dominating species in all the trials. Total Pb concentration was associated with F5 in trial 1 and 4. F1 was quite high in trial 2 and 5, but found to be decreasing rapidly in the course of composting and becoming not detectable at 20th day in trial 2, whereas it is decreasing from 10.8 to 2.8% of total concentration during the composting process. He et al. (2009) reported a decrease in exchangeable fraction in composting

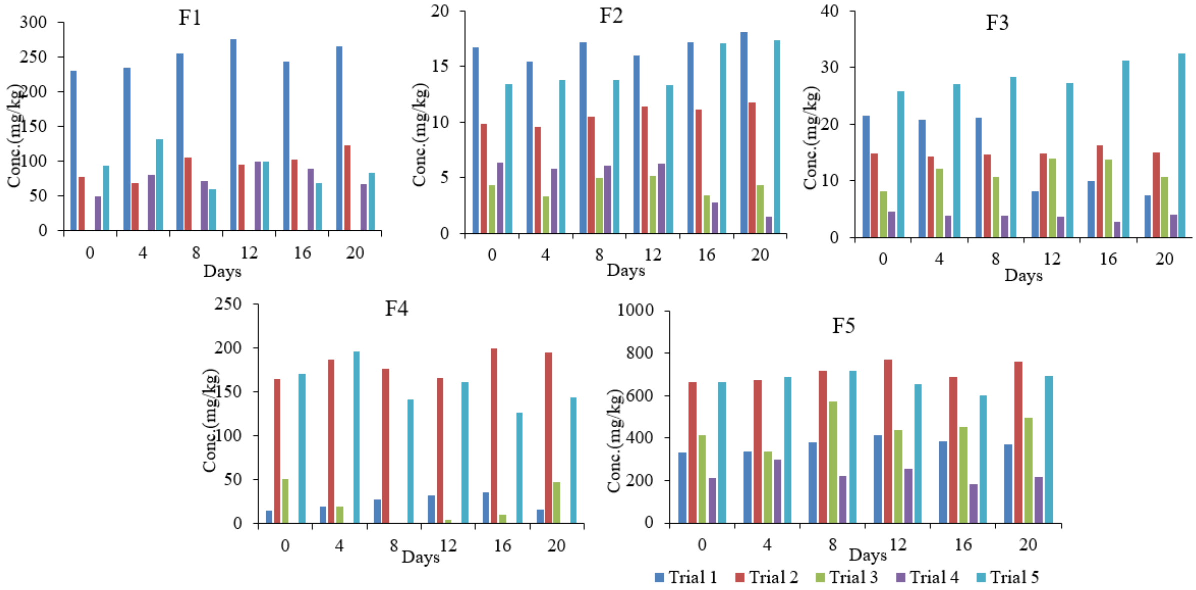

Fig. 2: Speciation of Cd in trial 1, 2, 3, 4 and 5 during the composting process.

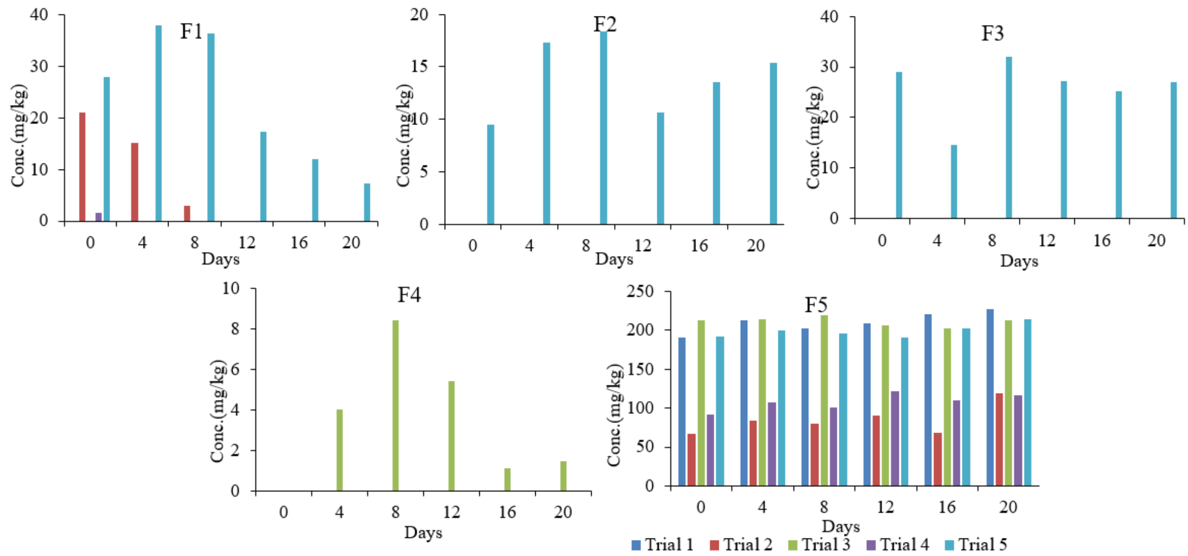

Fig. 3: Speciation of Pb in trial 1, 2, 3, 4 and 5 during the composting process.

of sewage sludge. The F2 fraction slightly increased but F3 decreased in trial 5. In trial 3, residual fraction is getting converted to oxidizable fraction. Singh & Kalamdhad (2012) also observed an increase in F2 fraction during water hyacinth composting. In trial 2, F5 sharply increased during the composting, whereas in trial 3 it got decreased. F5 increased from 74.2 to 81.2% in trial 5. Amir et al. (2005), He et al. (2009) and Singh & Kalamdhad (2012) have also reported F5 to be the most dominant concentration during composting and they also observed that the concentration is increasing with the progress in composting. BF of Pb rapidly went below detection limit in trial 2, while in trial 3 it slightly increased in thermophilic phase and then again got decreased and in trial 5, the value decreased from 0.25 to 0.18 in the course of composting. From these results we can see that possibility of Pb toxicity in plants is very less.

Table 2: Variation of different fractions of Cr during composting.

Days	Trial 1					Trial 2				
	F1	F2	F3	F4	F5	F1	F2	F3	F4	F5
0	17.72 ±0.32	16.72 ±0.83	21.42 ±0.11	8.2 ±0.74	56.37 ±1.86	12.44 ±0.12	9.87 ±0.59	14.75 ±0.5	14.45 ±0.45	55.24 ±0.61
4	16.87 ±0.03	15.47 ±0.26	20.75 ±0.53	11.34 ±0.25	64.24 ±1.36	11.85 ±0.23	9.6 ±0.43	14.34 ±0.67	13.77 ±0.28	47.27 ±0.31
8	16.87 ±0.03	17.14 ±0.35	21.09 ±0.68	7.35 ±0.25	70.8 ±1.9	14.19 ±0.38	10.5 ±0.22	14.7 ±0.19	14.62 ±0.06	54.9 ±0.88
12	17.57 ±0.21	16 ±0.58	8.22 ±0.35	5.7 ±1.81	67.2 ±0.52	13.09 ±0.06	11.42 ±0.08	14.79 ±0.21	14.24 ±0.36	51.4 ±0.44
16	17.37 ±0.51	17.15 ±0.31	9.95 ±1.48	7.84 ±1.03	64.94 ±1.01	13.09 ±0.29	11.15 ±0.06	16.2 ±0.05	15.19 ±0.33	56.94 ±1.46
20	18.64 ±0.31	18.1 ±0.4	7.4 ±0.94	4.9 ±0.1	71.3 ±4.03	12.95 ±0.33	11.79 ±0.28	14.94 ±0.41	14.69 ±0.13	59 ±2.39
Days	Trial 3					Trial 4				
	F1	F2	F3	F4	F5	F1	F2	F3	F4	F5
0	2.95 ±0.87	4.3 ±0.95	8.1 ±0.37	12.4 ±0.41	80.4 ±2.56	5.94 ±0.42	6.35 ±3.67	4.6 ±0.11	8.65 ±0.55	60.4 ±1.49
4	3.45 ±0.46	3.35 ±0.57	12.05 ±0.77	11.05 ±0.32	90.3 ±3.44	6.9 ±0.3	5.8 ±3.35	3.8 ±0.2	20.6 ±0.86	63.6 ±1.99
8	1.65 ±0.53	5 ±1.16	10.6 ±0.45	11.15 ±0.86	84.2 ±1.46	7.12 ±0.21	6.05 ±3.5	3.8 ±0.31	14.15 ±0.48	60.7 ±2.85
12	1.2 ±0.33	5.15 ±0.84	13.9 ±0.79	9.95 ±0.73	67.7 ±2.86	6.87 ±0.41	6.25 ±3.61	3.65 ±0.09	13.55 ±0.37	60.5 ±2.16
16	1.6 ±0.43	3.45 ±0.08	13.65 ±0.68	11.2 ±1.24	79.9 ±3.14	6.2 ±0.15	2.8 ±1.62	2.75 ±0.17	10.35 ±0.28	60.2 ±1.26
20	0.55 ±0.18	4.3 ±0.22	10.7 ±1.14	9.75 ±0.97	91.1 ±2.13	5.95 ±0.08	1.5 ±0.87	4.05 ±0.29	9.55 ±0.67	65.1 ±1.79
Days	Trial 5									
	F1	F2	F3	F4	F5					
0	2.99 ±1.17	13.45 ±0.38	25.89 ±0.8	30.42 ±0.86	117.84 ±2.34					
4	7.89 ±0.53	13.75 ±1.22	26.99 ±1.13	32.62 ±0.3	107.64 ±0.66					
8	8.5 ±1.13	13.82 ±1.11	28.27 ±0.47	36.19 ±0.64	132.77 ±0.84					
12	8.8 ±0.14	13.34 ±3.62	27.32 ±1.74	33.79 ±1.59	146.57 ±40.82					
16	12.74 ±2.03	17.07 ±2.11	31.29 ±0.66	35.04 ±1.64	112.3 ±2.43					
20	11.95 ±0.97	17.34 ±0.89	32.49 ±0.46	34.54 ±1.5	120.34 ±0.71					

(ND = not detected)

Chromium: Table 2 explains the variation in different fractions during speciation of PMS compost. In trial 1, 2 and 4, F1 fraction remained almost unchanged, but in trial 3 and 5 a decrease and increase was observed, respectively. Pare et al. (1999) observed a decrease in F1 fraction during composting of MSW. The F3 fraction also did not changed much during composting, except in trial 1 where it reduced from 17.8 to 6.2%. This reduction in F3 fraction may be caused due to the conversion of Cr (VI) to Cr (III) during the composting process. No distinct change was observed in F2 and F4 fractions in all the trials except in trial 4, whereas F2 fraction quickly decreased from 7.4 to 1.7%. The F5 fraction contributed the maximum to the total concentration. Liu et al. (2007) has also reported that F5 fraction was the dominating species during composting of sewage sludge. An increase in F5 fraction was observed in all the trials except trial 5, where it reduced from 61.8 to 55.5%. Singh & Kalamdhad (2012) observed a decrease in F5 fraction during composting of water hyacinth. Pare et al. (1999) has reported an increase in the F5 fraction during MSW composting. BF was found to be decreased in all the trials except trial 5. This increase in BF for Cr shows that presence of excess cattle manure could increase the mobility of chromium.

CONCLUSIONS

Chemical speciation is very valuable in assessing bioavailability of heavy metals during rotary drum composting of PMS. Residual fractions of all toxic metals were found to be the predominant fraction and also increased during PMS composting. The exchangeable and carbonate fractions were only detected for Pb in trial 5. It seems that addition of cattle manure in excess quantity is converting these metals to mobile fraction. An addition of cattle manure decreased the exchangeable and carbonate fractions of Cr during composting. Bioavailable fraction was quite high for Cd. A reduction in bioavailability factor of Cd and Pb was observed during composting in all the trials. It can be concluded that an addition of cattle manure in adequate quantity could change the physico-chemical properties of compost, resulting in influence to the distribution of different fractions of toxic metals during rotary drum composting of PMS.

ACKNOWLEDGEMENTS

The authors greatly acknowledge the financial support through the SRF award by the Council of Scientific and Industrial Research (CSIR), India and Department of Science and Technology (DST), Science and Engineering Board (SERB) Government of India for the necessary amenities vide Project No. SB/S3/CEE/0030/2013.

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