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Transformation of elemental toxic metals into immobile fractions in paper mill sludge through rotary drum composting

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ABSTRACT

Heavy metal content has become a serious issue with increase in compost application in agricultural land. The bioavailable fraction (water soluble and diethylene triamine pentaacetic acid (DTPA) extractable) is the major concern comparing to the total concentration as it can be taken up by plants which in turn affects the human health. Thus, this study was carried out to determine the variation in bioavailable and leachable fraction of heavy metals (Cd, Cu, Fe, Ni, Pb, Cr, Zn, Hg and Mn) and to assess the influence of temperature, pH, organic matter degradation and humification during rotary drum composting of paper mill sludge (PMS) for 20 days. A total concentration of all the metals as well as nutrients increased during composting. The Hg was not detected in water soluble and DTPA extractable fraction, whereas Pb and Cr were also not detected in water soluble fraction in all the trials. A decreasing trend was observed in leachable quantity for all the metals during composting. Water extractable quantity increased for Fe and Cu during composting. Organic matter degradation and humification during composting influences the variation in bioavailability and leachability of heavy metals, which can be optimized by applying appropriate proportion of cow dung during rotary drum composting of PMS.
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1. Introduction

Development of a society can be indexed by its paper consumption. Consumption of paper is increasing gradually due to development and industrialization. In the pulp and paper industry, Global manufacturing is predictable to increase by 77% by 2020 (Likon and Trebse, 2012). Approximately, 60 m^3 of wastewater is produced per tonne of paper production in the paper manufacturing process (Thompson et al., 2001). PMS is generated after the treatment of the effluent. The process adopted and wastewater treatment technology used are the major parameters influencing the production of sludge. Per tonne of paper production generally generates 40–50 kg dry sludge and it is a mixture of primary (70%) and secondary (30%) sludge (Bajpai, 2015).

In the near future, with the increase in paper production, proportional amount of sludge will also be produced indicating sludge management is of utmost concern. As of now, most of the sludge produced goes for landfilling. Scott and Smith (1995) reported that 69% of current production goes for landfilling, whereas 21% goes for incineration, 8% goes for landspreading and 8% goes for other alternatives. Due to presence of high amount of organic matter, the sludge will produce greenhouse gases (CO_2 and CH_4) in landfills. The economics of incineration is dubious as moisture content of PMS is around 30–50% whereas fibre content is approximately 30–50% of dry mass. In case of landspreading, there is a risk of chromium toxicity (Norris and Titshall, 2011). Other alternatives includes composting, using as landfill cover material, using for production of building material etc. Due to high organic content, composting is the most suitable recycling method for paper mill sludge. Phytotoxic effect of the raw sludge gets minimized after composting. It also kills or reduces pathogens. Composting can enhance the cation exchange capacity and nutrition retention potential of soil (Sesay et al., 1997), as well as it increases handling features (Das et al., 2001).

A PMS sludge rich in heavy metal concentration is becoming serious problem, these metals can cause serious environmental problems after land application (Singh and Kalamdhad, 2013a). Heavy metals are taken up by plants in small quantity for metabolism, but concentration higher than required becomes toxic (Carlson et al., 1975). Heavy metals are present in soil can affect the metabolism, which may hinders the growth and morphology of soil microbes, resulting loss of soil fertility (Bragato et al., 1998). It is not

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possible to predict about the bioavailable fraction of heavy metals from total concentration, though it can be used as an overall pollution indicator. The bioavailable portion is the main concern as it is taken up by the plants, which accumulates in the human tissue and biomagnifies through food chain, which can result in serious health and environmental problems (Singh and Kalamdhad, 2012). The bioavailability and eco-toxicity to plant is mainly dependent on their specific chemical bonding not on the total content of metals (Fuentes et al., 2006). The Toxicity Characteristics Leaching Procedure (TCLP) extractable concentration represent the leachable quantity of heavy metals in acidic condition. The DTPA extractable concentration also represents the plant available fraction of heavy metals.

Thus, the objective of this study is to assess the variation in bioavailable and leachable fraction of heavy metals during rotary drum composting of paper mill sludge.

2. Material and method

Primary Paper mill sludge (PMS), cow dung 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. 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 campus supplied the cow dung. Sawdust was bought from nearby saw mills. The bamboo chips were segregated and lumps of the PMS were broken to 1 cm particle size to facilitate better aeration. The proportions and initial characterization of raw materials is shown in Table 1.

For making compost out of PMS, a rotary drum composter (Capacity 500 L) was used as described by Singh and Kalamdhad (2013a). Proper degradation and stabilization of PMS was the key parameter in fixing the duration of composting as 20 days. Grab sampling was carried out for collection of samples from the various positions of the drum, mainly top, bottom and middle layer. 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. Triplicates samples were collected at each 4 day interval till the end of composting process. These samples were then milled and sieved through 0.2 mm sieve after drying for 24 h at 105 °C.

DTPA solution was prepared by mixing 0.005 M DTPA, 0.01 M $CaCl_2$ and 0.1 M triethanolamine and then pH was adjusted to 7.3. For TCLP, acetic acid with a pH of 4.93 ± 0.05 (adjusted with 1 N NaoH) was used. For digesting the sample for total metals, a mixture of H_2SO_4 and $HClO_4$ (5:1 V/V) was used.

Heavy metals were analyzed using Atomic Absorption Spectrophotometer (Varian Spectra 55B). A Flame Photometer (Systronics 128) was used for the analysis of micronutrients (Na, Ca and K). Total concentration of heavy metals were determined after the digestion of compost samples in a Block Digester (Pelican equipments Chennai, India) at 300 °C for 2 h. Temperature was measured using a digital thermometer (Mextech).

Water extract of compost was used to measure pH. Moisture content was determined by heating the sample at 105 °C for 24 h. The water-soluble heavy metals were extracted after shaking a solution of 2.5 g sample with 50 mL of distilled water at room temperature for 2 h in a shaker at 100 rpm. For extracting DTPA extractable concentration, 4 g powdered sample was mixed with 40 mL of DTPA reagent and shaked at 100 rpm. TCLP extractable metals were extracted by mixing 5 g of sample with 100 mL of acetic acid at room temperature for 18 h in a shaker at 30 ± 2 rpm. All the samples were then centrifuged followed by filtration and stored at 4 °C for further analysis.

Table 1
Initial Characterization of raw materials.

Parameter	Waste Material(Kg)		
	PMS	Cattle Manure	Saw Dust
Trial 1	150	0	0
Trial 2	120	15	15
Trial 3	105	30	15
Trial 4	90	45	15
Trial 5	75	60	15
pH	7.33 ± 0.05	6.8 ± 0.05	6.14 ± 0.05
Moisture Content(%)	46 ± 0.8	86.4 ± 0.9	9.6 ± 0.5
Volatile solids(%)	40.15 ± 0.5	72.05 ± 0.22	79.25 ± 0.23
Nutrients (g/kg)			
Na	2.938 ± 0.429	1.08 ± 0.01	2.5 ± 0.05
K	0.455 ± 0.036	0.7 ± 0.008	0.96 ± 0.02
Ca	31.56 ± 8.914	2.620 ± 0.02	9.89 ± 0.03
Total concentration(mg/kg)			
Cd	1600.5 ± 161.595	59.95 ± 0.5	52.63 ± 1.2
Cu	43.666 ± 1.527	29.5 ± 0.7	44.8 ± 0.5
Fe	3666.667 ± 813.9	2735.3 ± 11.3	1860.8 ± 2.8
Ni	89.5 ± 5.634	267.5 ± 1.5	239.9 ± 2.1
Pb	320.000 ± 25.980	1000 ± 5	847.55 ± 5.5
Cr	220.833 ± 35.809	93.27 ± 0.5	121.45 ± 0.5
Zn	23125.900 ± 1071	124.45 ± 2.25	188.4 ± 1.95
Hg	1883.333 ± 125.8	143.45 ± 2.5	195 ± 2.45
Mn	378.667 ± 14.978	173 ± 3.5	532.5 ± 1.5
Water extractable concentration(mg/kg)			
Cd	16.187 ± 2.846	ND	ND
Cu	2.100 ± 0.485	1.1 ± 0.02	3.2 ± 0.02
Fe	9.333 ± 5.441	163.98 ± 2.2	187.4 ± 2.5
Ni	0.687 ± 0.110	ND	ND
Pb	1.067 ± 0.231	ND	ND
Cr	3.180 ± 0.410	2.15 ± 0.6	5.12 ± 0.03
Zn	219.760 ± 11.227	6.1 ± 0.06	7.4 ± 0.04
Hg	41.133 ± 2.212	ND	ND
Mn	8.067 ± 0.436	5.205 ± 0.165	19.9 ± 0.13
DTPA extractable Concentration(mg/kg)			
Cd	25.187 ± 1.266	7.9 ± 0.6	11.6 ± 0.8
Cu	2.040 ± 0.075	3.6 ± 0.3	10.7 ± 1.7
Fe	90.203 ± 17.308	285.4 ± 2.5	216.2 ± 4.2
Ni	ND	ND	ND
Pb	3.027 ± 0.150	ND	ND
Cr	ND	14.8 ± 0.2	16.6 ± 0.6
Zn	192.127 ± 5.843	33.7 ± 0.2	52.4 ± 1.1
Hg	23.333 ± 1.528	ND	ND
Mn	61.613 ± 5.514	79.9 ± 0.2	274.3 ± 2.64
TCLP extractable Concentration(mg/kg)			
Cd	520.84 ± 23.726	59.4 ± 1.2	68.7 ± 0.9
Cu	17.547 ± 0.531	2.8 ± 0.02	3.1 ± 0.1
Fe	65.453 ± 1.630	25.6 ± 0.33	133.2 ± 0.6
Ni	12.760 ± 1.100	16.72 ± 0.4	25.7 ± 0.8
Pb	7.467 ± 0.306	11.4 ± 0.2	13.7 ± 0.3
Cr	21.400 ± 3.747	3 ± 0.145	4.04 ± 0.3
Zn	2366.67 ± 290.9	16.1 ± 0.5	18.5 ± 0.6
Hg	104 ± 6	9.2 ± 0.6	12.3 ± 0.4
Mn	126.947 ± 1.135	33.5 ± 0.5	51 ± 0.8

(ND–not detected; n = 3).

3. Results and discussion

3.1. Physico-chemical parameters

The temperature profile of the rotary drum composting of PMS is shown in Fig. 1. Trials with high amount of cow dung (trials 4 and 5) are showing a rapid increase in temperature due to high

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