

Evaluation of the Feasibility of Utilizing Composted Sludge from Leather Industry Wastewater in Sudan

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Abstract: *The objective of this study is to assess the potential for composting chromium-rich sludge from the leather industry wastewater in Sudan, with the aim of removing or capturing heavy metals before they can leach into the surrounding environment. "This research aims to evaluate the pH, organic matter, total nitrogen, phosphorus, potassium, and calcium content of compost made from leather industry wastewater sludge, as well as determine the level of bioavailability of heavy metals. To create the compost, the leather industry wastewater sludge was mixed with organic waste such as chicken manure, rice bran, and sawdust in various ratios, and then shaped into block piles for the composting process. The results indicate that for the recommended block-shaped piles, the organic content decreased from 30% to 25%, while the pH value and NPK content were satisfactory for composting. Additionally, the total heavy metal content, including Cr, Cd, Cu, Pb, Mn, and Zn, was well below the upper limit standard for biosolids, which classifies the compost as excellent." "The compost was found to be free of Salmonella sp., Shigella sp., and helminth eggs, and there was a 10³-fold decrease in total coliforms. The compost characteristics indicated that it was mature, and the germination index for cabbage seeds was 85.5%, which suggests that most of the phototoxic compounds were removed."*

KEYWORD: leather industry, Sludge, Heavy metals, Compost, Organic matter, Beneficial organism

INTRODUCTION

Various types of organic waste are now being explored and utilized as soil amendments for growing different plant species, including ornamental, food, and forest crops. These organic waste materials include yard waste composts, sewage sludge (Goldstein, 1991), and leather industry wastewater sludge, which is a by-product of leather manufacturing and serves as a source of

nitrogen. The leather manufacturing process produces three types of by-products: solid waste from splitting and trimming hides, sludge from liming, dehairing, pickling, and chrome tanning, and liquid waste from each step of the operation (Anderson,1981; Kabbashi, *et al.*, 2006). However, there is concern about the presence of harmful heavy metals such as polychlorinated compounds (Lisk *et al.*, 1992; Manos, *et al.*, 1992). Due to the diverse composition of leather industry wastewater sludge, there is no universally accepted approach for its management worldwide. Various solutions have been proposed, tested, and implemented at semi-pilot, pilot, and industrial scales." Previous studies have confirmed the successful stabilization of leather industry wastewater sludge in building materials such as bricks, concrete, tiles, and ceramics, as well as other engineering applications (Montañés *et al.*, 2014; Basegio *et al.*, 2002; Juel *et al.*, 2017). However, none of the processes mentioned above have been developed to a level that can fully meet industrial demands. Moreover, there is still a need for more attention to be given to treatment processes for this hazardous tannery sludge (Alibardi and Cossu, 2016).

Composting is considered as one of the alternative methods for converting organic waste into products that can improve plant growth and serve as soil amendments. The primary objective of composting is to produce a stable compost product that contains adequate nutrients for plant growth and can enhance soil fertility." Cellulose and lignin are two primary components of agro-based biomass that are considered as main sources of energy and humus formation, respectively. Additionally, their characteristics contribute to air permeability, bulking, and water retention during the composting process (Hubbe *et al.* 2010). While extensive research has been conducted on composting using various organic wastes (Baharuddin *et al.*, 2009a; Hock *et al.*, 2009; Heerden *et al.*, 2002; Khalil *et al.*, 2001), there is limited information available on field-scale composting of leather industry wastewater sludge." The aim of this study is to examine the physical and chemical characteristics, as well as microbial succession, during the composting process of leather industry wastewater sludge with chicken manure and rice bran. The specific objective is to assess the role of chicken manure in enhancing the degree of degradation and improving the quality of the end product."

Material and Methods

Preparation of Materials

The composting process was conducted at the composting site located in the Khartoum tannery in Khartoum, Sudan. The composting treatment was carried out using brick blocks with dimensions of 2.0 m in length, 2.0 m in width, and 2.0 m in height. The composting blocks were placed under shade and on a cement base. The fresh leather industry wastewater sludge was collected from a closed anaerobic digesting tank system located at the Khartoum tannery wastewater treatment

plant in Khartoum, Sudan. The collected sludge was then dried and ground into small sizes using a conventional mill machine at the same location.

Composting Process

A composting block was constructed using approximately 200 kg of composting materials with a ratio of 6:3:2:1 of leather industry wastewater sludge, rice bran, chicken manure, and sawdust, respectively. The composting pile was placed under shade on a cement base. Beneficial microorganisms from the anaerobic tannery sludge were sprayed onto the composting pile every four days using a centrifugal pump. The composting material was manually turned to ensure good mixing and aeration after the addition of anaerobic tannery sludge. The addition of anaerobic tannery sludge was stopped ten days before the compost reached maturity and followed by frequent turning. Samples were collected systematically at the end of each week, air-dried for 10 days, and then ground into a fine powder. The maturity stage of the compost was determined every four days by analyzing the C/N ratio using a CNHS 2000 analyzer (Leeco, USA).

Sampling and Analysis Techniques

Samples were collected weekly from each composting block at three different depths (top, middle, and bottom) using a stainless-steel corer with a diameter of 5 cm. The samples were stored in sterile plastic bags and immediately transported to the laboratory for analysis.

Physical and chemical analysis

The temperature was measured using a thermometer, while moisture content was determined by the gravimetric method. pH was measured using a pH meter (Hanna HI 991001, Romania), and electrical conductivity was determined by using a conductivity meter (Eutech Instruments, Singapore) (Hafidi *et al.*, 1994). Total nitrogen content was determined using the Kjeldahl method. Organic matter content was analyzed using the loss-on-ignition method. The C/N ratio was determined using the CNHS 2000 analyzer (Leeco, USA).

Microbial analysis

Microbial analysis was carried out by standard plate count techniques using Nutrient Agar, Sabouraud Agar, and MacConkey Agar for total bacterial count, fungal count, and total coliform count, respectively. The most probable number (MPN) technique was used to determine the fecal coliform count. The identification of microbial isolates was carried out using standard microbiological and biochemical tests (Al-Othman *et al.*, 2012).

Statistical analysis

The differences among several mixtures were estimated using the one-way ANOVA method. Additionally, the relationship between different concentrations and chemical parameters was evaluated using Pearson's correlation coefficient. The statistical analysis was performed using the SPSS program.

RESULTS AND DISCUSSIONS

Properties of Materials

The tannery sludge has an alkaline pH of 7.450 ± 0.04 and over 83% of it is made up of particles smaller than 50μ , giving it a silty clay texture. It is also rich in organic matter, but contains twice as much calcium, sodium, and nitrogen as typical soil (Table 1). The physical and chemical properties of the samples used in this research study showed significant differences ($p < 0.01$). The pH showed noticeable improvement ($p < 0.05$) and a strong correlation ($r = 0.93$) with the cumulative concentration of the tannery sludge. Table 1 presents the characterization of the sludge from leather industry wastewater. The sludge has a low C/N ratio (38.12) and a high nitrogen content (0.80). However, the leather industry wastewater sludge has high levels of heavy metals, including Cr, Cd, and Pb, which may have negative effects on plant growth. The Cr level is very high (1550 ± 2.9 mg/l), exceeding the permissible level in soil (100 mg/kg) (Chaney and Ryan, 1993; Chaney, 1990a). The low solubility of Cr allows only a small amount of it to be bioavailable (Alloway, 1990), which means that phytotoxicity is rarely observed even when crops are grown in soils treated with sludge high in chromium. The leather industry wastewater sludge also has a high Pb content (95.0 ± 3.90 mg/l), exceeding the maximum level in soil (15 mg/l) (Chaney and Ryan, 1993; Chaney, 1990a). Cadmium, copper, iron, manganese, and zinc all have satisfactory concentrations, with levels of 3.4 ± 1.03 , 39.6 ± 1.05 , 750.0 ± 8.90 , 40.6 ± 2.03 , and 42.4 ± 4.30 ppm, respectively (Table 1). Overall, the leather industry wastewater sludge has satisfactory concentrations of these metals, except for Cr, Cd, and Pb (Table 1).

Table 1. Physicochemical composition of the raw materials utilized in this study

Parameters	Tannery Sludge	Chicken Manure	Rice bran	Sawdust
Organic Carbon, %	30.5±0.56	25.5±0.75	40.35	50.46±5.01
Total Nitrogen, %	0.80±0.01	3.0±0.08	1.30	0.9±0.01
Potassium, %	0.48±0.90	1.8±0.05	1.50	0.08±0.03
Phosphorus, %	0.40±0.01	2.55±0.03	0.50	0.25±0.01
Calcium, %	5.50±1.30	1.45±0.04	1.00	0.05±0.01
Magnesium (ppm)	890±10.90	265±5.9	240.20	0.02±0.01
Iron (ppm)	750.0±8.90	950±8.5	200.30	350.46±7.1
Sodium (ppm)	450.0±5.50	85.0±2.9	90.80	65.0±3.9
C/N ratio	38.125±5.35	8.50±1.7	31.04	56.06±0.61
pH	7.450±0.04	7.25±0.07	8.45	8.25±0.07
EC (mS/cm)	3.68±0.05	5.35±0.20	6.45	1.35±0.20
Heavy Metals (ppm)				
Chromium (ppm)	1550±2.9	16.6±2.9	7.5	12.8±1.9
Cadmium (ppm)	3.4±1.03	0.7±0.01	0.3	5.4±0.3
Copper (ppm)	39.6±1.05	125.8±4.9	35.44	4.6±0.5
Manganese (ppm)	40.6±2.03	25.65±1.08	35.40	5.6±1.9
Lead (ppm)	95.0±3.90	1.45±0.06	1.3	16.4±2.3
Zinc (ppm)	42.4±4.30	123.6±4.9	130	8.8±1.3

values as mean standard error. SE= standard error

During the composting process, changes in color and texture were observed, with the final compost (matured) having a grayish color and an earthy texture similar to normal compost. Table 1 shows that sawdust can be used as a carbon source, and the chicken manure used in this study had high levels of nitrogen, potassium, and phosphorus, consistent with the results reported by other researchers (Baharuddin *et al.*, 2009a; Hock *et al.*, 2009). The addition of chicken manure (a nitrogen source) to the leather industry wastewater sludge compost can enrich and accelerate the composting process by providing a nitrogen source and microbial seeding, according to Hock *et al.* 2009.

The wet bulk density was close to that of mineral soil but when dry it is relatively light with bulk density equal to 0.14 g/cm (Table 2). The water holding capacity, bulk density, and pore space of the sludge were high when compared with the typical soils which have low ability to keep water for a long time in spite of their low bulk density and total pore space (Table 2).

Table 2. Physical properties of wastewater sludge of the leather industry utilized in this study

Parameters	Typical soil	Sludge (S)
Water retention at pressure (kPa) (%w/w, dry basis):		
0	33.40	153.41
1	32.20	117.50
10	22.80	89.50
33	21.20	78.50
1500	15.90	69.57
Available water	5.3	8.93
Bulk density (g/cm ³)	1.3	0.14
Total pore space	-	94

Evaluation of Nutrient Content in Compost Materials

The analysis of compost derived from leather industry wastewater sludge is presented in Table 3. The values for nitrogen (N), phosphorus (P), and potassium (K) were 98.60 ± 8.9 , $93.50 \pm 7.$, and $1,520 \pm 6.7$ ppm, respectively, but these nutrient elements may not be readily available to plants in organic form (He *et al.*, 2000). The calcium concentration in the compost ($69,840 \pm 15.7$) was high and exceeded the permissible level (30,000), but calcium-deficient acid soils can negatively affect crop quality, making compost a potential solution to increase calcium availability for crop growth in such conditions.

Table 3. Concentration of inorganic element in leather industry wastewater sludge, compost, and the maximum allowable limits as described by USEPA,1995 [21]

Trace element	Tannery sludge (ppm)	Tannery sludge compost (ppm)	USEPA limits,1995 (ppm)
Total Nitrogen, %	66.58 ± 2.9	98.60 ± 8.9	NM
Potassium, %	30.74 ± 3.3	80.99 ± 5.4	2.0 - 1,600
Phosphorus, %	8.46 ± 1.7	93.50 ± 7.9	2.7 - 400
Calcium, %	$77,000 \pm 10.7$	$69,840 \pm 15.7$	30,000
Magnesium (ppm)	$1,160 \pm 7.7$	$1,520 \pm 6.7$	530
Iron (ppm)	$1,062 \pm 7.4$	254 ± 6.7	5,000
Sodium (ppm)	$1,006 \pm 8.7$	$2,514 \pm 11.7$	NM
C/N ratio	38.125 ± 50.35	25.234 ± 5.35	15-25
pH	7.450 ± 0.04	8.450 ± 0.06	NM
EC (mS/cm)	3.6	9.5	4.8

The heavy metal content of the compost was below the threshold for excellent biomaterials classification (Table 4) (USEPA, 1995), likely due to dilution and losses through drainage, as sawdust and chicken manure were added to the compost.

Table 4. Comparison of Heavy Metal Concentrations in Leather Industry Wastewater Sludge and Compost with Maximum Allowable Limits by USEPA, 1995

Trace element	Tannery sludge (ppm)	Tannery sludge compost (ppm)	USEPA limits,1995 (ppm)
Chromium (ppm)	1550±2.9	80±2.7	1,200
Cadmium (ppm)	3.4±1.03	1.6±0.03	39
Copper (ppm)	39.6±1.05	54 ±1.7	1,500
Manganese (ppm)	40.6±2.03	20±2.03	NM
Lead (ppm)	95.0±3.90	3.2±0.7	300
Zinc (ppm)	42.4±4.30	148±0.7	2,800

Properties of the Matured Compost

The pH profile of the composting process is depicted in Figure 1. The initial pH of the compost was low and gradually increased over time. The pH fluctuated during the process and showed a slight drop towards the end. The rate of decomposition during composting is known to increase with increasing pH within the range of 6.25-8.5. The pH changes during composting are predictable. Initially, the pH may drop slightly due to oxygen limitation caused by fermentation. However, as the composting process progresses, the pH can increase up to approximately 8.4 due to ammonification. Once ammonification is complete, the pH will decrease to about 7.5-7.8 (Miller,1993).

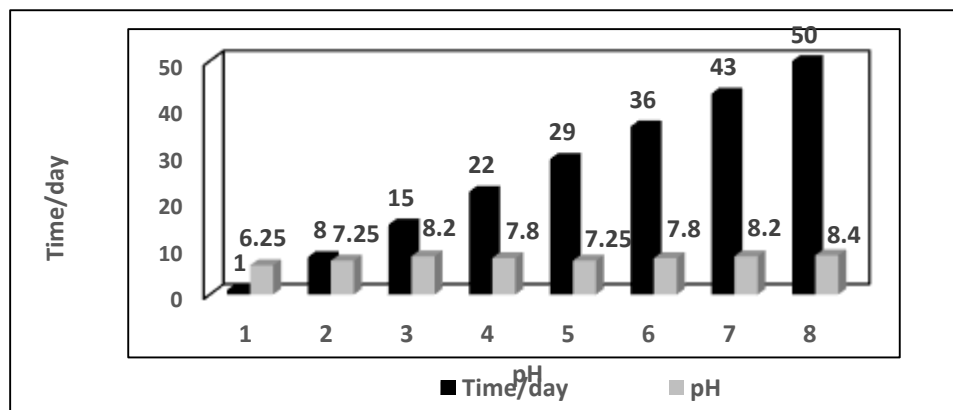


Figure 1. pH profile during the composting process

The carbon-nitrogen ratio of the compost decreased from 40 on the first day to around 25 after day fifty of the composting process, indicating a declining pattern (Figure 2). This decline was attributed to the formation and loss of carbon dioxide. During composting, some of the biodegradable carbon in the material is assimilated by the microbes and converted to microbial protoplasm, while the rest is oxidized by the microorganisms to carbon dioxide to meet their energy requirements. The carbon dioxide then diffuses into the surrounding air, leading to a reduction in the carbon content of the composting mass (Golueke,1973).

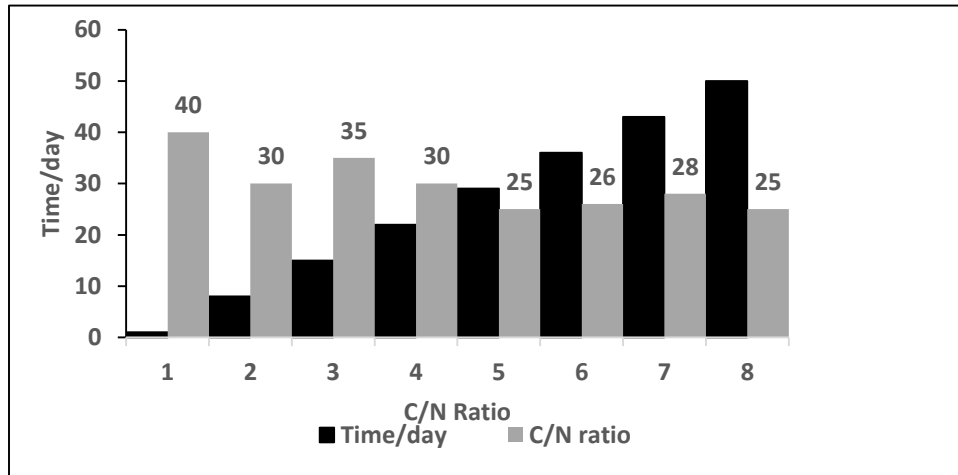


Figure 2. Changes in the C/N ratio during composting process

The electrical conductivity of the compost showed a significant increase ($p < 0.05$) and a strong positive correlation ($r = 0.97$) with the cumulative concentration of leather industry wastewater sludge, ranging from 48.5% to 73.3% in different samples (Figure 3). The EC value of 9.5 mS/cm was higher than the value of 4.8 mS/cm obtained by Van Heerden *et al.* 2002 in citrus supplement with calcium hydroxide composted for two months. However, despite the high EC value, the activity of the microbes during the composting process was not affected, as evidenced by the production of carbon dioxide. High EC values in compost may be attributed to the presence of microbial cells (Brock *et al.*, 1994). It has been reported that salt concentrations above 8 mS/cm can have a negative impact on microorganism populations and the biotransformation of organic matter (Santamaria and Ferrera, 2001).

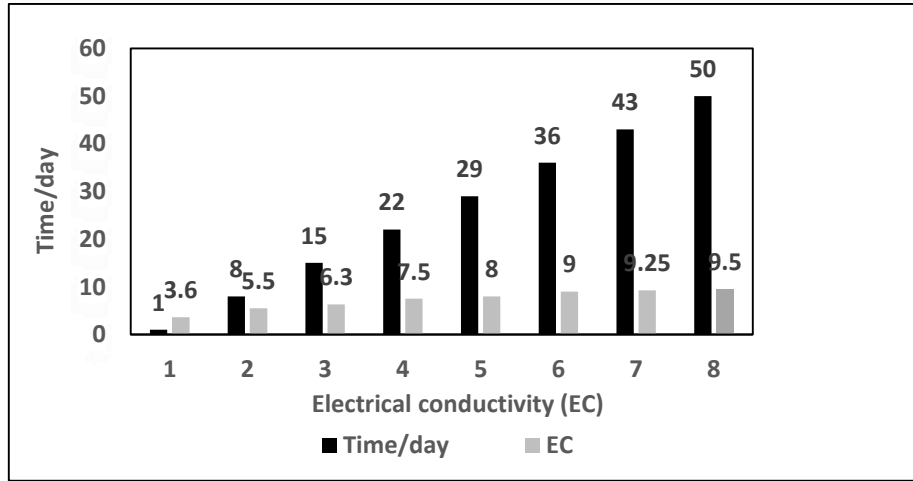


Figure 3. Electrical conductivity (EC) during composting process

The compost temperature profile showed an increasing pattern up to 53.5°C, followed by a decline to 35°C, which marked the end of the composting process (Figure 4). Leather industry wastewater sludge compost has been reported to reach high thermophilic temperatures due to its high organic content. The compost content should be large enough to maintain high temperatures within the composting block, where the width is typically between 5-8 meters, and the height can be up to 2-2.5 meters.

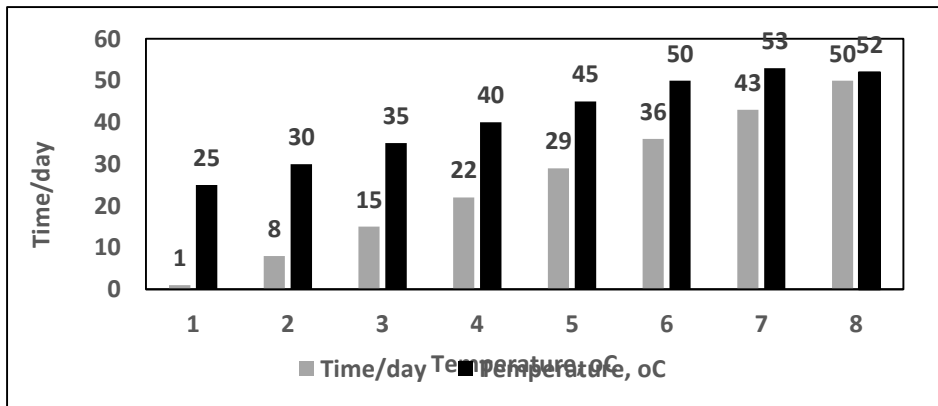


Figure 4. Temperature Variation throughout composting process

The high temperature (55.5°C) reached during composting was sufficient to reduce beneficial organisms, as total coliforms decreased significantly, while no *Shigella* sp., *Salmonella* sp., eggs of helminths, or fecal coliforms were detected (Table 5). Some studies have shown that the solid waste of the sewage industry can be decontaminated through composting, and the beneficial organism content may drop below detectable levels before the 27th day of composting (Deportes *et al.*, 1998).

Table 5. Beneficial organism in the compost components and maximum allowed limits as stipulated by USEPA, 1999.

Microorganism	Tannery sludge	Chicken manure	Compost	USEPA Limits
Faecal coliforms	< 10	< 10	< 10	< 10 ⁵
Total coliforms	2 × 10 ⁷	2 × 10 ⁶	2 × 10 ⁴	NM
Helminthes eggs	ND	ND	ND	< 10 × 10 ⁴
<i>Shigella</i> sp.	ND	ND	ND	< 5
<i>Salmonella</i> sp.	ND	ND	ND	NM

Assessment of compost toxicity:

The compost toxicity was evaluated by determining the germination index using an aqueous extract of the compost, and a value of 85.5% was obtained. A germination index value above 50% is generally considered to indicate satisfactory maturity of the compost (Mathur *et al.*, 1993a, b). However, the presence of phytotoxic compounds, such as acetic, propionic, butyric, and isobutyric acid, that have not been metabolized can inhibit germination (Epstein, 1997).

CONCLUSION

The leather industry wastewater sludge was combined with organic waste such as chicken manure, rice bran, and sawdust to enhance its content and reduce its phytotoxicity. Through the composting process, pathogens and helminth eggs were reduced, resulting in the production of stable and mature compost with a germination index suitable for use with Chinese cabbage. The total concentration of heavy metals in the compost remained within allowable limits and complied with the USEPA 1995 standard, making it a suitable fertilizer and soil conditioner.

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Disclosure of conflict of interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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