

# **leachability and Bioavailability Approaches of Heavy Metals During Composting of leather Industry Wastewater Sludge**

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## **Abstract**

Varied operational strategies of composting blocks were used in this study to assess the phytotoxicity. Five composting blocks that varied in composting approaches (i.e., turning frequency and moisture content adjustment) were made. Composting of biomass of rice straw by adding suitable quantity of chicken manure and rice husk enhanced the degradation of microbial along with the decreasing of the heavy metal's bioavailability of the composting blocks. Moisture content varied between 35-16%, while volatile solid escalating between 35-13.3% in blocks 2 & 5 respectively. In the same manner the higher and lower declining in water-soluble and leachable metals was observed in block 2&5. On the other hand, pH in all composting blocks was approached the neutral status. With regard to heavy metals the total content of nickel, lead, and zinc in the composting blocks were higher compared to the total content of chromium; but the water-soluble concentration of chromium in the composting blocks were relatively little bit high compare to that concentration of water-soluble nickel, lead, and zinc which means the high chromium noxiousness. The content of leachable heavy metals of the composting blocks were within the permeable levels. Alteration the biomass of rice straw into composting materials may service in the protection of environment and meanwhile reduce the utilization of non-environmentally friendly fertilizers (chemical fertilizers).

## **Keyword:**

Heavy metals, water-soluble, leather industry, Composting blocks, phytotoxicity,

## **1. Introduction**

The bioaccumulation of heavy metals over huge area of land and long time periods, may result in accumulation and the gradual damage to ecosystem, requires careful monitoring of the input, mobility, and effects of these pollutants [1,2,3]. One of the human activities which effect global balance is leather industry wastewater sludge, a by-product of leather manufacturing. The leather industry generates large volumes of effluents with high concentration of dyes and salts [4]. The

discharge of leather industry wastewater sludge into the environment without appropriate treatment poses a serious threat for the ecosystem. Tanning industries generates wastes with different physical and chemical properties. The main tanning method for production of hard leather (sole leather) is a vegetable tanning, while chromium tanning is for production of light, smooth, and soft leather suitable for working goods such as shoe uppers, handbags, gloves and a variety of clothes. Though several handling measures for setting tanning by-products have been suggested, sludge from leather industry wastewater depuration is still an environmental problem. This study discovers the possibility of recycling leather industry wastewater sludge into a soil reclamation by utilizing suitable composting techniques. Composting has been considered as one of the alternative methods to convert organic wastes into products that benefit plant growth and soil amendment. The major goal of composting is to provide a stable compost product that contains sufficient nutrients to be consumed by the plant and also can increase soil fertility. Two of the main components of agro-based biomass, i.e., cellulose and lignin, have been described as main sources of energy and humus formation, respectively, and their characteristics also contribute to air permeability, bulking, and water retention throughout the composting process [5]. Although considerable research on composting has been conducted using various organic wastes [6,7,8,9], there is less information regarding leather industry wastewater sludge composting at the field scale of operation. Thus, the goal of the current research work was to determine the bioavailability, leachability, and differences of total nutrients and heavy metals during composting of the Rice Straw modified with leather industry wastewater sludge, chicken manure, and rice husk.

## 2. Material and Methods

### 2.1 Preparation of Raw Materials

Rice Straw was collected from Agricultural field and brought to the composting shed near Khartoum tannery- Khartoum-Sudan. Chicken manure and rice husks were obtained from Soba farms -Khartoum Sudan. The composting materials was prepared by reducing the sizes to 15 mm using different methods (cutting/shredding) and mixing with Chicken manure and rice husk. Table 1& 2 designate the quantities of the composting materials and the physico chemical properties of the five different blocks used in this study.

**Table 1.** Composting blocks contents (kg)

Parameters	Rice straw	Chicken Manure	Rice Husk
Block 1	30	60	10
Block 2	45	45	10
Block 3	70	20	10
Block 4	75	15	10
Block 5 (control)	100	0	0

**Table 2.** Characteristic of the composting materials

Parameters	Tannery Sludge	Chicken Manure	Rice husk	Rice straw
pH	3.5±0.56	6.6±0.75	6.12±0.05	5.8±0.7
EC (mS/cm)	6.4±0.05	3.8±0.08	1.60±0.01	5.5±0.01
Moisture content, %	45.5±1.90	78.4±0.9	10.50±0.9	85.5±1.90
Volatile solid (VS), %	3.4±0.03	70.4±0.33	63.9±0.65	68.5±0.55
Potassium (mg/kg)	4200±6.2	9056 ± 22.5	9520 ± 15.4	19,870 ± 87.0
Calcium (mg/kg)	7700±18.5	9831 ± 45.5	4320 ± 15.6	8273 ± 70.0
Magnesium (mg/kg)	1190±4.4	5795 ± 35.5	995 ± 9.8	7545 ± 65.0
Sodium (mg/kg)	1006±6.2	3200 ± 8.5	1920 ± 6.3	6129 ± 25.5
<b>Heavy Metals (ppm)</b>				
Chromium (mg/kg)	1404±0.99	135.6 ± 0.8	15 ± 0.5	155.9 ± 2.35
Cadmium (mg/kg)	3.23±0.03	39.9 ± 0.9	57 ± 0.50	43.9 ± 3.5
Copper (mg/kg)	41.7±0.63	48.7 ± 0.8	25 ± 0.35	43.8 ± 0.90
Iron (mg/kg)	1062±3.03	1972 ± 8.5	3980 ± 33.4	6544 ± 35.5
Nickel (mg/kg)	125±1.03	266± 3.5	142± 2.4	268±5.5
Manganese (mg/kg)	70±0.03	1026 ± 11.5	450 ± 5.3	1082 ± 10.0
Lead (mg/kg)	93±0.77	765 ± 0.9	55 ± 0.23	747± 0.85
Zinc (mg/kg)	44.67±0.55	183.4 ± 1.95	102 ± 2.34	215 ± 0.50

## 2.2 Composting Process

The materials of compost were designed into composting blocks, that consisted of approximately 500 kg of composting materials of leather industry wastewater sludge, Rice husk, Chicken manure, and rice straw respectively. The five different composting blocks containing were composted for thirty days and sampling was carried out after manual turning on 0, 5, 10, 15, 20, 25, and 30<sup>th</sup> days. Three different samples were collected randomly from the middle & bottom of the composting block making one kilogram and mixed carefully to form a homogenous mixture. Collected samples were immediately air dried at 105 °C in oven, ground to pass through 0.2 mm sieve and stored for the subsequent chemical and physical analysis.

### 2.2 Physico-chemical analysis

A digital thermometer was used to measure temperature during the composting process. The gravimetric method by weight loss of the wet sample of compost was used for determination of Moisture content [10]. A pH and a conductivity meter were used to assess the pH and electrical conductivity (EC) of the composting samples on the filtrate (Whatman filter paper no. 42). Volatile solids (VS) were determined by using Ignition method (550 °C for 2 h in muffle furnace). Ammonium acetate procedure was used to determine the total content of Calcium,

Sodium, Potassium, and Magnesium. Manganese, Chromium, iron, cadmium, copper, nickel, and lead were determined by using atomic absorption spectrometer (Varian Spectra 55B) [10]. Extraction method by using 2.5 g sample with 50 mL of distilled water at room temperature for 2 h in a shaker at 100 rpm were used for estimation of water-soluble nutrients and heavy metals [11]. USEPA Method 1311 was used for estimation test of Toxicity characteristic leaching procedure (TCLP) [12].

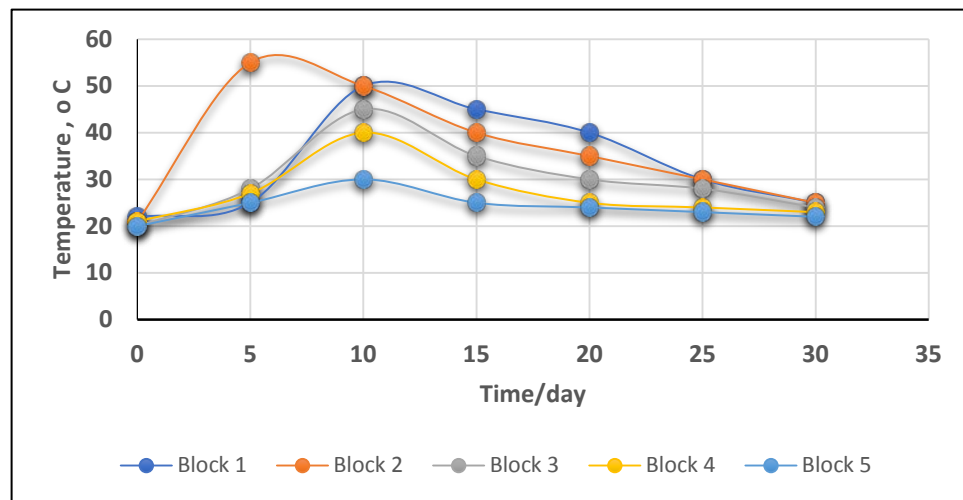
## 2.4. Statistical analysis

ANOVA method of one way have been used to estimate differences among several mixtures and means of three replicates. running statistical analysis was done by SPSS program.

## 3. Results and Discussions

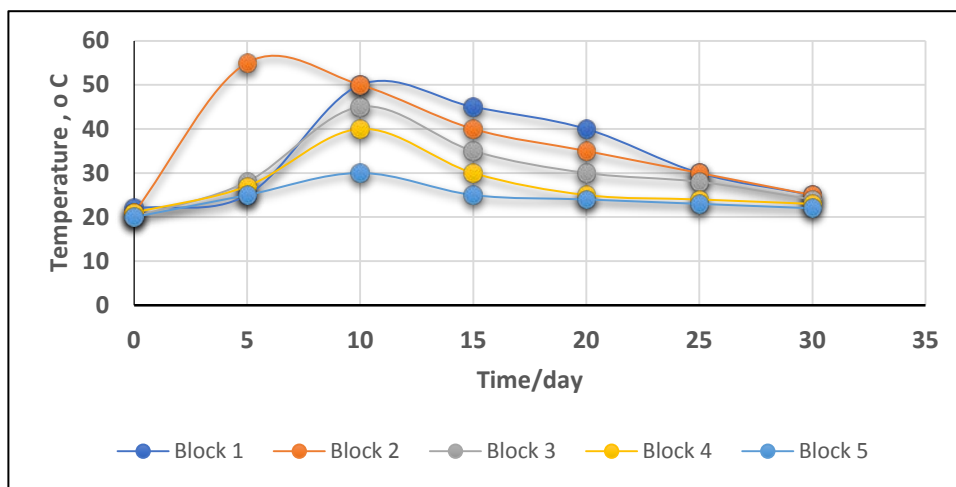
### 3.1 Properties of Raw Materials

Figure 1 shows the temperature profile during the course of composting. The temperatures of the blocks show increasing pattern within the first three days of the process. The peak temperature (55 °C) was measured on the fifth day in block two whereas the block one approached 50 °C on the seventh day. Blocks three& four displayed high temperatures of 45 and 40 °C on the 10<sup>th</sup> day of the composting process, respectively. The control block 5 showed peak temperature of only 30 °C on the 7th day of the composting indicating that block composting of control without addition of cattle manure and rice husk was not feasible. The rise of temperature through all blocks owing to the release of heat caused by microbial catabolism. Some researcher reports reflect the decontamination of solid waste of sewage industry through the process of compost and discovered that the content of beneficial organism was declined to below the recognition limits before the 27<sup>th</sup> day of composting [13].



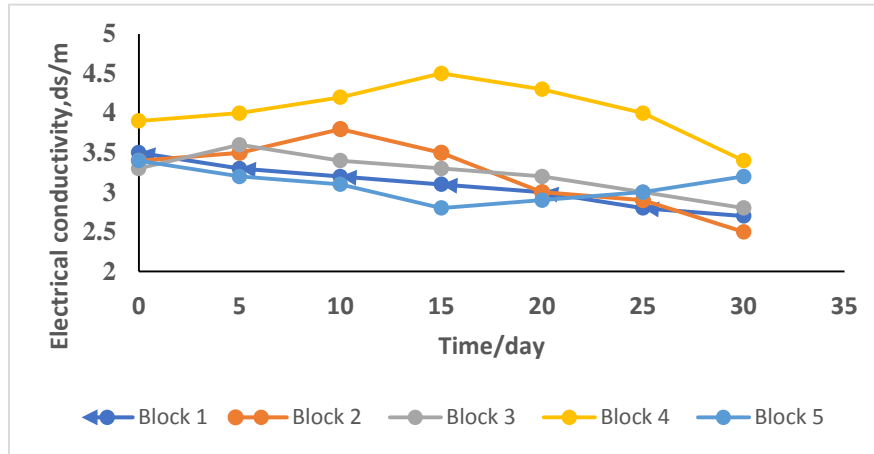
**Figure 1.** Temperature trend of the block during composting process

According to the previous studies composting displayed no more than 60-80% moisture content during the course of the process [14]. The moisture content should be at low level for survival of beneficial organisms, whereas high moisture content decreases the rate of oxygen diffusion through the pores. During the course of composting moisture content is scaped to atmosphere as water vapor, and the reduction in moisture content indicated by degradation index rate [15]. The loss in moisture content in the blocks is in the following sequences: block 2 (35%), block 1 (26.25%), block 3 (22.5%), block 4 (18%), and block 5 (16%) respectively (Figure 2). Therefore, block two had the higher degradation frequency which was coordinate with the trend of temperature of the block. Moisture content in all the blocks were generally above 60 % during the active phase. Therefore, the variation of moisture content between the blocks is significantly important ( $P < 0.05$ ).



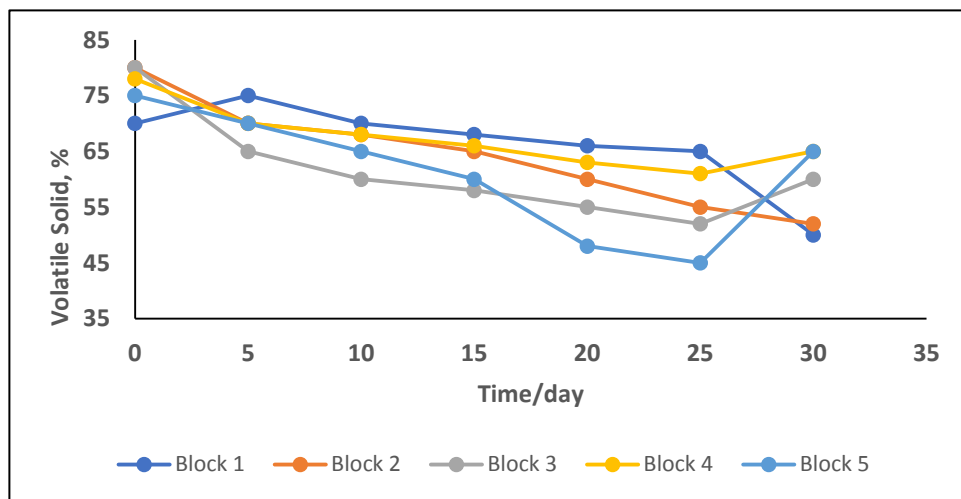
**Figure 2.** Moisture content variation of the block during composting process

Figure 3 display the electrical conductivity in which there is a partial increase in the electrical conductivity in all composting blocks owing to the production of mineral salts such as ammonium & phosphates ions during organic matter degradation [16]. Electrical conductivity was reduced in block one, two, three, & four this owing to the decomposition of ammonia compound as volatile materials and deposition of mineral salts [17]. Blocks 1, 2, 3, & 4 in the end of compost processing showing electrical conductivity of 2.7, 2.5, 2.8 & 3.4 dS/m respectively. Previous studies proof ed that the degree of salinity is affected by the electrical conductivity [18]. One of the limitations of utilization of compost as fertilizers is its low capability of intake of water of plants owing to high concentration of salts that increase the effect of osmosis [19]. Generally electrical conductivity of 4 dS/m or above in the final composts will adversely affect plant growth, e.g., low germination rate, withering, etc. [20,21,22]. Hence, composts with high electrical conductivity must be mixed with other materials like soil having low electrical conductivity before applying for crop growth. Electrical conductivity among our studied blocks was significantly important ( $P < 0.05$ ).



**Figure 3.** Electrical conductivity variation of the block during composting process

Volatile solids normally represent extent of organic matter in waste, compost, wastewater, and have great importance in water and wastewater treatment. A common scenario for volatile solids that it declines thru the composting process owing to organic content degradation and release of carbon in the form of carbon dioxide by the beneficial organisms. In this research work, block 2 approach the maximum decline of volatile solids (35 %), thus the trends of declining of volatile solids were in the order block 2 (35%)> block 1 (28.6%) > block 3 (25%)> block 4 (16.6%)> block 5 (13.3%) (Figure 4). Declining concentration of composting blocks of volatile solids were significant ( $P > 0.05$ ).



**Figure 4.** Volatile Solid variation of the block during composting process

Table 3 shows the whole content of nutrients (sodium, potassium, calcium, & magnesium) and heavy metals (chromium, cadmium, copper, iron, nickel, manganese, lead, & zinc) throughout the course of the composting process. The total concentrations of both nutrients and heavy

metals increased during the composting owing to degradation of organic matter indicated by reduction of weight resulting in release of carbon dioxide and the subsequently mineralization [23]. The trends of increasing of nutrient is as follows Potassium>Calcium>Magnesium>Sodium> while the order of arrangement of the heavy metals is Iron>Manganese >Lead >Nickle> Zinc> Chromium> Copper > Cadmium. The rise of content of heavy metals in the soils owing to extended utilization of composts which might cause harmfulness to humans, plants, and animals. Consequently, the study of bioavailability of the nutrients and heavy metal contents in the plants are vital before spread on the compost to the agriculture field.

### **3.2 Heavy metals and nutrients Bioavailability**

All composting blocks shows an increase in water soluble content of the nutrient (Table 4). The potassium (K) and calcium (Ca) and magnesium (Mg) exhibit lower solubility in block five which contained only rice straw biomass. The rise in content of water-soluble nutrients was similarly owing to reduction of dry matter weight during the degradation and mineralization of the organic matter [24]. Therefore, the dissimilarity in concentration of water-soluble nutrients between the composting blocks was significantly ( $P<0.05$ ).

Heavy metals belong to the most harmful constituents and its water solubility considered most toxic fraction in the compost [25,26,27]. All the composting blocks exhibit significant decreasing pattern ( $P<0.05$ ) in regard of water-soluble concentration of heavy metals.

On the other hand, cadmium and nickel water-soluble fraction were not detected in all the composting blocks. The decreasing pattern of water-soluble forms of Zinc, Copper, Manganese, Iron, Lead and Chromium throughout the composting process may be due to the binding of the metals with the hydroxyl and carboxylic acid groups augmented by chicken manure. So, these organic groups and the fresh formed humus raised the combined sites and joint with metals to form insoluble and immobile complexes [23,27,28]. Previous studies confirmed the increase in iron water -soluble concentration [29], but in the current research decreasing of water-soluble content of iron in all composting blocks was noticed.



**Table 3.** Total nutrient and heavy metals content of the initial and final mature compost

	Total nutrient concentration mg/kg dry matter									
	Block 1		Block 2		Block 3		Block 4		Block 5	
	initial	final	initial	final	initial	final	Initial	final	initial	final
Sodium	5896±75	9856±75	5620±55	10856±75	5856±45	7856±43	5856±23	7634±34	6123±56	7231±55
Potassium	16432±98	19456±86	17532±51	17532±51	16456±86	20456±86	18564±23	20156±23	19756±23	5856±23
Calcium	8432±85	10432±89	8532±45	12532±49	7532±51	9532±54	7832±56	10532±58	8234±45	10462±49
Magnesium	6132±58	8432±69	6432±60	9324±59	6124±64	7346±56	6524±67	8124±76	7543±58	7332±55
	Total heavy metals concentration mg/kg dry matter									
	Block 1		Block 2		Block 3		Block 4		Block 5	
	initial	final	initial	final	initial	final	Initial	final	initial	final
Chromium	97.5±2.2	170±6.4	87.5±2.6	150±4.3	80.6±2.6	147±4.5	110±4.6	200±6.2	155±8.2	220±8.7
Cadmium	48.5±0.26	59.1±0.21	48.2±0.20	58.4±0.22	43±0.15	55.4±0.27	50±0.13	59.4±0.24	44±0.8	60.1±0.25
Copper	50.7±0.23	90.1±0.28	62.7±0.33	94.1±0.30	51±0.35	88.4±0.33	46±0.30	86.4±0.32	44±0.7	83.2±0.34
Iron	6100±58	8430±69	5610±36	7450±66	6423±55	8130±64	7654±45	9865±78	6543±43	7620±59
Nickle	276±8.4	345±5.6	272±4.7	327±3.9	282±4.5	303±3.6	291±4.5	347±3.6	267±3.5	303±2.8
Manganese	810±8	1020±15	1082±22.7	1620±25.6	1184±26	1590±28	1180±33	1480±32	1080±31	1140±23
Lead	760±1.8	830±2.5	740±2.3	835±3.4	710±2.8	805±3.7	730±3.8	809±2.7	748±0.9	840±3
Zinc	198±1.5	220±2	160±0.9	210±1	200±2.9	235±2	207±2.4	224±2.2	215±2.8	254±3.6

Mean ± SD (n = 3) (ANOVA: P<0.05)



**Table 4.** Water- soluble of nutrient and heavy metals content of the initial and final mature compost

	<b>Water soluble nutrient concentration mg/kg dry matter</b>									
	<b>Block 1</b>		<b>Block 2</b>		<b>Block 3</b>		<b>Block 4</b>		<b>Block 5</b>	
	<b>initial</b>	<b>final</b>	<b>initial</b>	<b>final</b>	<b>initial</b>	<b>final</b>	<b>Initial</b>	<b>final</b>	<b>initial</b>	<b>final</b>
Sodium	1520±15	1860±20	1640±17	1960±30	1600±14	1620±25	1630±14	1560±25	1650±14	1480±25
Potassium	2340±45	4309±39	2120±44	4410±43	2820±44	4459±43	2886±44	4059±43	3009±40	4160±34
Calcium	2348±15	3420±65	2148±15	3520±65	2548±15	3720±65	2648±25	3420±15	2748±12	3390±19
Magnesium	1876±45	2750±29	1776±20	2950±26	1976±17	2750±22	1976±17	2750±22	7243±46	6832±54
	<b>Water soluble heavy metals concentration mg/kg dry matter</b>									
	<b>Block 1</b>		<b>Block 2</b>		<b>Block 3</b>		<b>Block 4</b>		<b>Block 5</b>	
	<b>initial</b>	<b>final</b>	<b>initial</b>	<b>final</b>	<b>initial</b>	<b>final</b>	<b>Initial</b>	<b>final</b>	<b>initial</b>	<b>final</b>
Chromium	34.5±0.22	25.6±0.19	39.5±0.16	29.5±0.09	39±0.16	32.2±0.09	37±0.16	32.4±0.09	38±0.13	33.2±0.07
Cadmium	-	-	-	-	-	-	-	-	-	-
Copper	8.3±0.15	5.6±0.28	7.3±0.10	5.0±0.09	8.1±0.09	5.5±0.07	8.4±0.09	6.5±0.07	8.7±0.15	7.2±0.12
Iron	250±0.10	140±0.30	258±0.19	130±0.20	268±0.9	180±0.20	238±0.8	150±0.25	268±0.18	210±0.29
Nickle	-	-	-	-	-	-	-	-	-	-
Manganese	61.5±0.12	40.9±0.15	66.5±0.11	47.9±0.13	66.2±0.9	50.9±0.10	71.2±0.9	54.9±0.10	65.4±0.12	46.7±0.15
Lead	9.8±0.16	7.6±0.23	11.5±0.14	7.8±0.17	9.5±0.12	6.8±0.15	11.3±0.12	9.6±0.11	10.2±0.12	9.4±0.10
Zinc	11.5±0.08	8.6±0.12	11.2±0.08	8.6±0.06	15.2±0.8	11.6±0.09	15.2±0.09	10.3±0.07	16.1±0.06	12.4±0.09

Mean ± SD (n = 3) (ANOVA: P<0.05)

**Table 5.** Leachable concentration of heavy metals content during composting process

	<b>Leachable heavy metals concentration (mg/kg dry matter)</b>									
	<b>Block 1</b>		<b>Block 2</b>		<b>Block 3</b>		<b>Block 4</b>		<b>Block 5</b>	
	<b>initial</b>	<b>final</b>	<b>initial</b>	<b>final</b>	<b>initial</b>	<b>final</b>	<b>Initial</b>	<b>final</b>	<b>initial</b>	<b>final</b>
Chromium	70.5±5.22	50.6±3.19	79.5±6.24	35.6±2.23	59.5±4.1	41.6±3.2	62.5±4.1	46.6±3.2	67.5±5.3	55.6±3.3
Cadmium	1.9±0.32	0.88±0.03	2.53±0.04	1.22±0.06	2.8±0.06	1.71±0.03	2.64±0.04	1.88±0.05	2.69±0.04	2.03±0.07
Copper	22.3±0.30	16.2±0.28	26.4±0.50	16.2±0.28	26.6±0.5	18.1±0.1	26.6±0.5	19.1±0.1	25.6±0.5	19.0±0.1
Iron	470±2.09	310±1.30	495±2.10	350±2.30	405±2.08	397±3.30	475±3.08	377±3.08	470±2.88	397±4.11
Nickle	180±2.09	135±1.05	198±1.09	142±0.9	210±0.9	157±0.7	202±0.5	174±1.05	206±0.8	182±1.15
Manganese	319±3.09	250±1.50	413±5.09	244±4.50	403±4.19	237±3.50	405±4.22	250±5.09	407±6.22	264±2.09
Lead	58.5±0.5	39.6±0.9	65.7±0.9	41.6±0.8	58.5±0.7	41.4±0.8	67.5±0.9	50.4±1.09	66.1±0.8	57.4±0.9
Zinc	125.2±0.4	99±0.5	124.3±0.2	96±0.3	142±0.5	125±0.6	137.3±0.5	117±0.01	129.2±0.7	120±0.3

Mean ± SD (n = 3) (ANOVA: P<0.05)

### **3.3 Toxicity characteristic leaching procedure (TCLP) test for heavy metals**

Table 5 displayed variation of heavy metals leachability such chromium, cadmium, copper, iron, nickel, manganese, lead, and zinc during the course of composting. All the composting blocks exhibit reduction pattern of leachability during the composting of rice straw in the range of 17.6-55.2% for chromium; 24.5–53.7% for cadmium; 25.8–38.6% for copper; 15.5–34.04% for iron; 11.6–28.3% for nickel; 21.6–41.2% for manganese; 13.2–36.7% for lead; and 7.1–22.8% for zinc. Heavy metals leachability during the composting process was also reported by so many researchers [30,32,33,34]. The maximum allowable level for heavy metals' contamination is: Chromium (100 mg/kg), Cadmium (20 mg/kg) and lead (100 mg/kg) [15]. Therefore, findings of Toxicity characteristic leaching procedure (TCLP) test proofed that the heavy metal contents in composting blocks were below the permeable boundaries for compost utilization in agriculture. Decreasing of heavy metals leachability throughout the composting process may be attributed to the complexity formed with humus substances [28,30]. pH also had played a vital role in the decreasing of this leachable phenomena of the metals. Complexation, soluble, and surface legends were controlled by pH, while increasing of pH within the optimal range lead to reducing the solubility and bioavailability of heavy metals [35]. Compost Statistical estimation exhibit the important variances in the leachable concentration of heavy metals of all composting blocks ( $P < 0.05$ ).

## **4. Conclusion**

Bioavailability decreasing of compost is done by the optimal addition of rice straw with the chicken manure, and rice husk that enhanced the microbial degradation as well as leachability of heavy metals of the composting materials. Moisture content varied between 35-16%, while volatile solid escalating between 35-13.3% in blocks 2 & 5 respectively. In the same manner the higher and lower declining in water soluble and leachable metals was noticed in block 2&5. On the other hand, pH in all composting blocks was approached the neutral status. With regard to heavy metals the total content of nickel, lead, and zinc in the composting blocks were higher compared to the total content of chromium; but the water-soluble concentration of chromium in the composting blocks were relatively little bit high compare to that concentration of water-soluble nickel, lead, and zinc which means the high chromium noxiousness. Heavy metals leachable contents were within the permeable levels. Alteration the biomass of rice straw into composting materials may service in the protection of environment and meanwhile reduce the utilization of non-environmentally friendly fertilizers (chemical fertilizers). Therefore, in case of application of optimal composting condition such as weekly adjustment of moisture to 60% & rotating of block every 4 days, maturation time of the compost may strongly proportional with phytotoxicity vanishing.

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