Jundishapur J Health Sci. 2018 July; 10(3):e78891. doi: 10.5812/jjhs.78891.

Published online 2018 August 1. **Research Article**

Feasibility Study of Chromium Removal from Paint Sludge with Biological Sludge, Using Vermicompost by *Eisenia fetida* (Case Study: Saipa Automotive Industry)

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Received 2018 May 15; **Revised** 2018 July 08; **Accepted** 2018 July 21.

Abstract

Background: During the paint spraying process in the automotive industry, a variety of wastes, including a large amount of paint sludge is produced, which contains a variety of hazardous substances, such as BTEX and heavy metals. The aim of the present study was vermicomposting of paint sludge with sewage sludge and *Eisenia fetida* as earthworm spices for reduction of heavy metals.
Objectives: Vermicomposting process employing earthworms is increasingly being used for conversion of both sewage and some industrial wastes to reduced heavy metals and volatile solid content in bulk of solid wastes. Therefore, in this study, feasibility of chromium removal from paint sludge by using *Eisenia fetida* worms was investigated.
Methods: In this study, the quantitative and qualitative characteristics of paint and biological sewage sludge waste were determined in terms of Cr^{+6}, moisture content, nitrogen and carbon, C/N ratio, pH and temperature. Then the paint sludge was mixed in various proportions with the thickening biological sewage sludge of Saipa's sewage treatment plant. The decomposition time for the vermicompost samples for each run was 90 days. In order to analyze various parameters, including Cr^{+6} and volatile solids reduction, sampling from each pilot was performed on 1, 15, 30, 50, 70 and 90 days. The Cr^{+6} measurements were performed using the AASM method. The paint sludge and sewage sludge were mixed in different ratios. After the mixing, 500 adult *Eisenia fetida* earthworms were introduced to each box.
Results: The results showed that the optimum removal rate for Cr^{+6} was found in the mixing ratio of 6 kg of biological sludge with 600 g of paint sludge and in this ratio, the amount of Cr^{+6} concentration fell from 6 mg/kg to less than 0.2 mg/kg in 90 days. In addition, in the same ratio of sludge mixing, the C/N ratio ranged from about 27 on the first day to 14.3 on day 90 and the pH decreased from 7.8 on the first day to 7.3 on the 90th day. Volatile solids decreased from 80.4% on the first day to 37% on the 90th day.
Conclusions: Vermicomposting is a good alternative for treating paint sludge with biological sludge, and is advantageous for reduction of pollutants purposes. Research was carried out on bioavailability of heavy metals such as Cr^{+6} during vermicomposting for 90 days using *Eisenia fetida* earthworms. Finally, the results showed that, *Eisenia fetida* could eliminate Cr^{+6} in low levels.

Keywords: Chromium, Automotive Industry, Biological Sludge, Paint Sludge, Vermicompost

1. Background

During the paint spraying process in the automotive industry, waste materials, such as a large amount of sludge (about 3 kg of paint sludge per car) are generated, and this sludge contains a variety of toxic compounds, including BTEX and heavy metals. Paint sludge is a hazardous waste because it contains various organic, mineral, and toxic contaminants, such as BTEX, and heavy metals, such as lead, cadmium, nickel, mercury, arsenic, etc., which, in addition to causing environmental pollution (i.e., water and soil) endanger human health. These compounds are widely used in petroleum products and as fuel additives to increase the octane number. The BTEX combinations are used as solvents in various industries, such as automotive, paint, adhesives, lubricants, etc. Heavy metals, by creating multiple mechanisms, cause different disorders and side effects among creatures .Some important disorders and side effects of heavy metals are: Carcinogenicity, central nerves system disorders, skin disease, hematopoietic system disorders, cardiovascular system problems, kidney damages, and accumulation in tissues (1-4). Some heavy metals, such as chromium and cadmium are allocated to group A1 (carcinogen) by the environmental protection agency, thus the maximum level in landfills is different for each element. This amount for chromium XI is 0.5 mg/L. Also, the world health organization suggested the maximum level for the mentioned metals in drinking water. This level for chromium XI is 0.05 mg/L (5-7). There are numerous methods for heavy metals elimination, such as burning, encapsulation, and solidation. Most current treatment technologies for heavy metals elimination are not economical. Therefore, nowadays biological treatments are suggested. According to the importance of this method, in the current study, paint sludge treatment by using vermicompost, was studied. Vermicompost is the result of a semi-aerobic process, which is done by special species of worm, fungus, bacteria, and actinomyces. Vermicompost consists of worms residue plus decomposed organic matters and worms body, which is valuable for plants and fertilized soil. There are four important kinds of worms, which are involved in fixing wastes: *Eisenia fetida, Dendrobaena veneta, Lumbricus rubellus, Lumbricus terrestris*. *Eisenia fetida* is the most used in producing vermicompost, which is seen in more than 27 sites in Iran. *Eisenia fetida* is from the Lumbricidae species. This worm is also named red worm, tiger worm, fertilizer worm, and compost worm. For the first time, the worm was found in Europe, Siberia, South Russia, Palestine, and North America (8-10).

Vermicompost is the result of a semi aerobic process, which is done by special species of worms, fungus, bacteria and actinomyces (11, 12). Vermicompost consists of worm's residue plus decomposed organic matters and worms body, which is valuable for plants and fertilized soil. Vermicompost is a bioorganic fertilizer, which is very soft, light, brittle, clean, odorless, and is similar to coffee granules. The worms accumulate in rich masses of organic matter and using them and their excrements cause soil quality improvements, and they tolerate different environmental situations (12-14). Their population increases by increasing accessible food. They work all year and need little care. Certain species of earthworms, such as *Eisenia fetida, Aporrectodea tuberculata, Lumbricus terrestris, L. rubellus, Dendrobaena rubida, D. veneta, Eiseniella tetraedra, Allolobophora chlorotica and Pheretima* spp. have been found to remove heavy metals, pesticides and lipophilic organic micro-pollutants, such as Polycyclic Aromatic Hydrocarbons (PAH) from soil. Mining activities unleash large amounts of different heavy metals and contaminate vast land areas. They routinely give out arsenic, cadmium, lead, mercury, nickel and beryllium (14-17). Earthworms can tolerate high concentrations of heavy metals in the environment. Various researchers have reported that earthworms can bio-accumulate high concentrations of metals, including heavy metals, in their tissues without affecting their physiology and this particularly occurs when the metals are mostly non-bioavailable. Studies indicate that earthworms can take up and bio-accumulate heavy metals, such as cadmium (Cd), mercury (Hg), lead (Pb) copper (Cu), manganese (Mn), calcium (Ca), iron (Fe) and zinc (Zn). They can particularly ingest and accumulate extremely high amounts of heavy metals. Cadmium levels up to 100 mg per kg dry weight have been found in tissues. Of all the metals, Cd and Pb appear to accumulate in most species of earthworms at greater level. Earthworms collected from roadsides and mining sites show higher amounts of heavy metals than those from other sites. Thus, earthworms can also work as a 'bio-indicator' of heavy metal contamination in soil (11, 18-23). The effective factors in producing fertilizer by vermicomposts (*Eisenia fetida*) are C/N ratio. Furthermore, pH, chemical compounds, salt percentages, oxygen levels, particles salt, volatile solid percentage, and earthworms accumulate on the surface. The main goal of this study was to measure lead and zinc metals concentration in SAIPA automotive industry painting wastes, and eliminate them by using vermicompost with sewage sludge. In this study, heavy metals, such as Pb and Zn concentration reduction rate was analyzed before and after the vermicomposting process (5, 21, 24) .

The vermicomposting is used by previous researchers for the conversion of industrial sludge to vermicompost, for instance Gajalakshmi et al. (25) used paper mill sludge, Bansal and Kapoor (26) used agricultural waste, Sinha et al. (27) used domestic kitchen waste, and Elvira et al. (22) used paper mill sludge (6). Previous literature shows that vermicomposting of industrial sludge by using *Eisenia fetida* was useful for the utilization of industrial sludge as a vermicompost for agricultural purposes (21, 22, 25, 28).

2. Objectives

The aim of the present study was vermicomposting of paint automotive industries sludge using *Eisenia fetida* earthworm species to convert to usable sewage sludge, obtain an optimum composition of the industrial sludge and sewage sludge for various different compositions of feed materials, and study the variation of physico-chemical parameters for different compositions of feed mixture. Therefore, the general objective of this study was to measure the concentration of Cr^{+6} and Volatile solids in paint sludge of Saipa Automobile Company and to reduce and remove them using the vermicompost process along with the biological sludge at Saipa's sewage treatment plant.

3. Methods

In this study, paint sludge containing hazardous waste, such as BTEX and heavy metals, were originally prepared from Saipa automobile paints. Due to the presence of BTEX, heavy metals, and other toxic compounds, the process of composting and vermicomposting by *Eisenia fetida*

Table 1. Characteristics of Pilots of Vermicompost Paint Sludge and Biological Sewage Sludge

Pilot	The Percentage of Biological Sludge	The Percentage of Paint Sludge	The Amount of Biological Sludge	The Amount of Paint Sludge	The Amount of Chromium Mg
A	50	50	6	6	49
B	67	33	6	3	26
C	80	20	6	1.5	16
D	86	14	6	1	10
E	89	11	6	0.750	6
F	91	9	6	0.600	5
G	94	6	6	0.375	4

worms with pure paint sludge wastes alone was not possible. Therefore, the sewage sludge of the waste water treatment plant was used with a high percentage of biodegradable organic matters at Saipa industry. To achieve this aim, a series of plastic (finely woven) mesh boxes weighing 20 kg in nine groups of three were used to determine how volatile solids and heavy metals, such as Cr^{+6}, are removed from fresh paint sludge combined with the sewage sludge of the sewage treatment plant using the vermicompost process. The mixture of the sludge was placed separately at different weight ratios of biological sludge mixed with paint sludge in the pilots (ratios of 1 to 1, 1 to 2, 1 to 4, 1 to 6, 1 to 8, 1 to 10, and 1 to 16). In order to access this point, 27 netting boxes, which are about 20 kg, were chosen. Paint and biological sludge mixture ratios were 1/1, 2/1, 4/1, 6/1, 8/1, 10/1, and 16/1. Firstly, paint sludge was crushed in mills and then, paint and biological sludge was mixed and placed in boxes, then about 500 worms were added to boxes. Samples were aerated slowly for three days per week. Samplings was done on the 1^{st}, 15^{th}, 30^{st}, 50^{th}, 70^{th} and 90^{th} day (in 90 days). As the C/N ratio is the main factor for starting the process and because of the low level of C/N ratio at the beginning of the process (about 25), some straw was used for increasing the ratio. Sampling was done after complete mixing and achieving the required weight (about 250 gr sample with 2 mm diameter), on specific days (1^{st}, 15^{th}, 30^{th}, 50^{th}, 70^{th}, and 90^{th} days). Physical and chemical characteristics of paint sludge and biological sludge were sampled and analyzed three times, separately. During the decomposition process, chromium amount, pH, moisture, dry solid sludge percentage, volatile solids percentage, carbon, nitrogen, C/N, and temperature were measured. For measuring the mentioned parameters, the standard methods were used and chromium was measured by atomic absorption spectrophotometry (17-19). Table 1 shows the characteristics of vermicompost pilots of paint sludge and sewage sludge.

4. Results

Qualitative properties of the waste used in the vermicompost process: Physical and chemical properties of paint sludge and sewage sludge of Saipa Automobile Industry Sewage Treatment Plant for the vermicompost process, according to Tables 2 and 3.

Table 2. Characteristics of Paint Sludge and Sewage Biological Sludge Used in the Vermicompost Process

Parameter	Paint Sludge	Biological Sludge
PH	8.25	6.8
Humidity	20%	65%
Dry solids	80%	35%
Organic or volatile solids	72%	88%
Stable or mineral solids	28%	22%
Nitrogen (mg/kg)	465	2.7
Carbon (mg/kg)	35100	15.2
C/N	75	15.6

Table 3. Carbon and Nitrogen Analysis and C/N Ratio at Start in Bulk of Solids in Vermicompost

Treatment	Organic Carbon	Nitrogen	C/N
6 kg/6 kg	30.93	3.965	7.8
6 kg/3 kg	29.7	3.96	7.5
6 kg/1.5 kg	28.08	3.9	7.2
6 kg/1 kg	26.22	3.8	6.9
6 kg/750 gr	23.63	3.75	6.3
6 kg/600 gr	22.94	3.7	6.2
6 kg/375 gr	21.6	3.6	6.00
Biological sludge	19.8	3.5	5.65
Pain sludge	35.1	0.465	75.5

Figure 1 shows the C/N ratio variations for all vermicompost pilot of paint sludge and biological sludge during decomposition of 90 days. As shown in the diagram, the amount of C/N ratio decreases with increasing time in the vermicomposting process.

Figure 2 shows the variation in pH during decomposition of 90 days of paint sludge and biological sludge by the vermicompost process. Biological sludge's pH level was in the neutral range (6.8) and paint sludge's pH level was in the alkaline range (8.6), which changed during the vermicompost process. Table 4 shows the volatile solids percentage variations for all vermicompost pilot of paint sludge and biological sludge during decomposition of 90 days. As

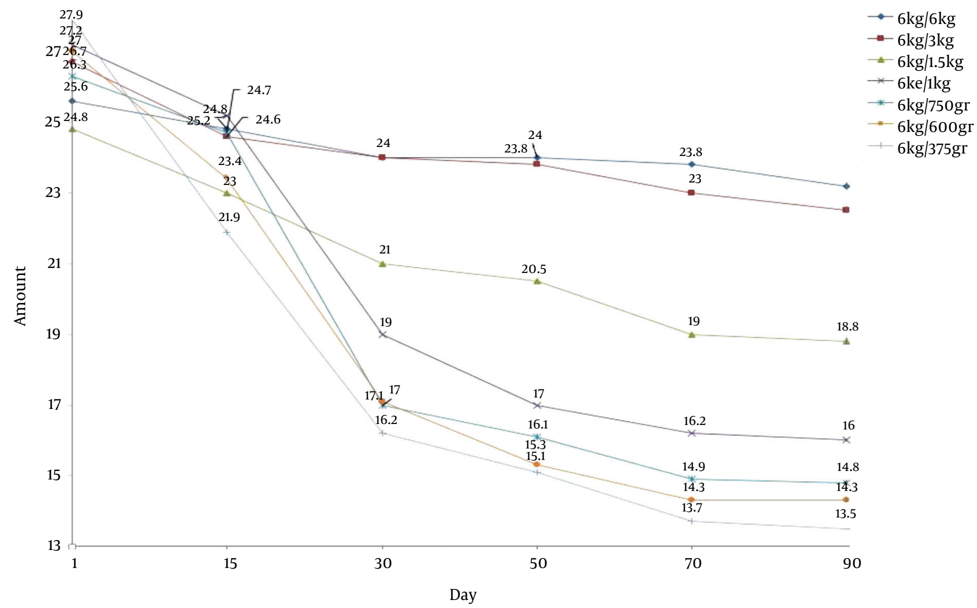

Figure 1. C/N ratio for all vermicompost pilot of paint sludge and sewage biological sludge

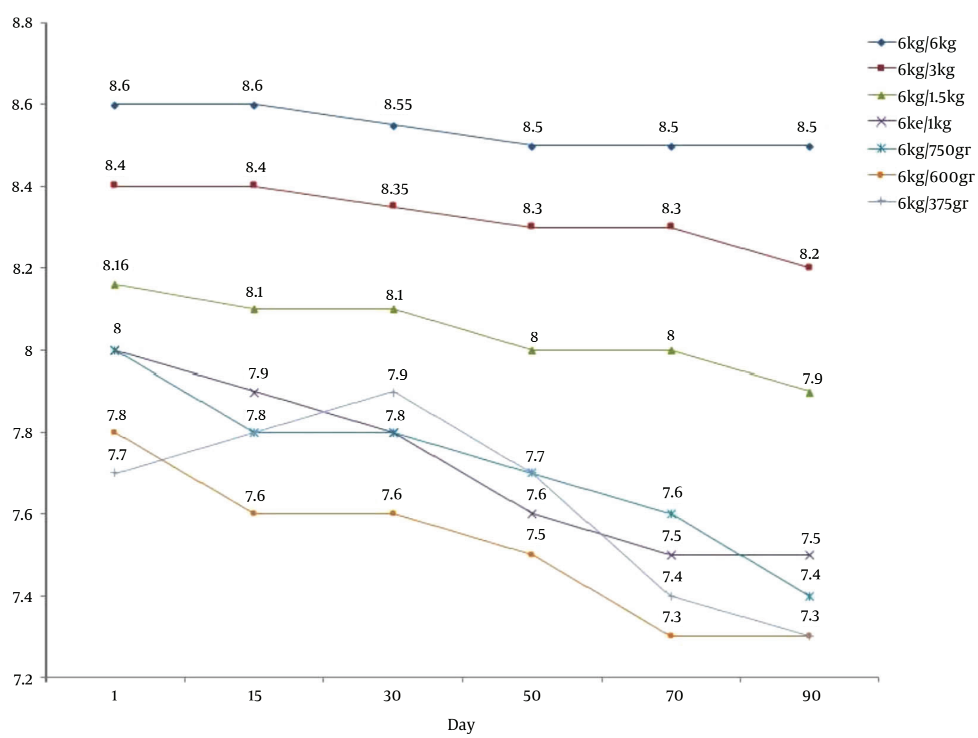

Figure 2. pH changes for all vermicompost pilot of paint sludge and sewage biological sludge

shown in the table 4, the amount of volatile solids percentage decreased with increasing time in the vermicomposting process.

Figure 3 shows the amount of chromium for all vermi-

Table 4. Reduction of Volatile Solids Percentage in Bulk of Vermicompost Sewage Sludge with Paint Sludge in mg/kg

Parameters	Time/Treatments	1 Day	15 Days	30 Days	50 Days	70 Days	90 Days
	6 kg/6 kg-A	60.6	57	55	53	53	53
	6 kg/3 kg-B	65	61	58	51	49	49
	6 kg/1.5 kg-C	72.8	64	60.5	52	43	42.4
The percentage of volatile solids	6 kg/1 kg-D	75	64.38	58.5	51.6	43	42
	6 kg/750 gr-E	78.3	65	56	51	41	40.7
	6 kg/600 gr-F	80.4	66	54.5	49	38.7	37
	6 kg/375 gr-G	86.3	68	51	44.5	36	34.5

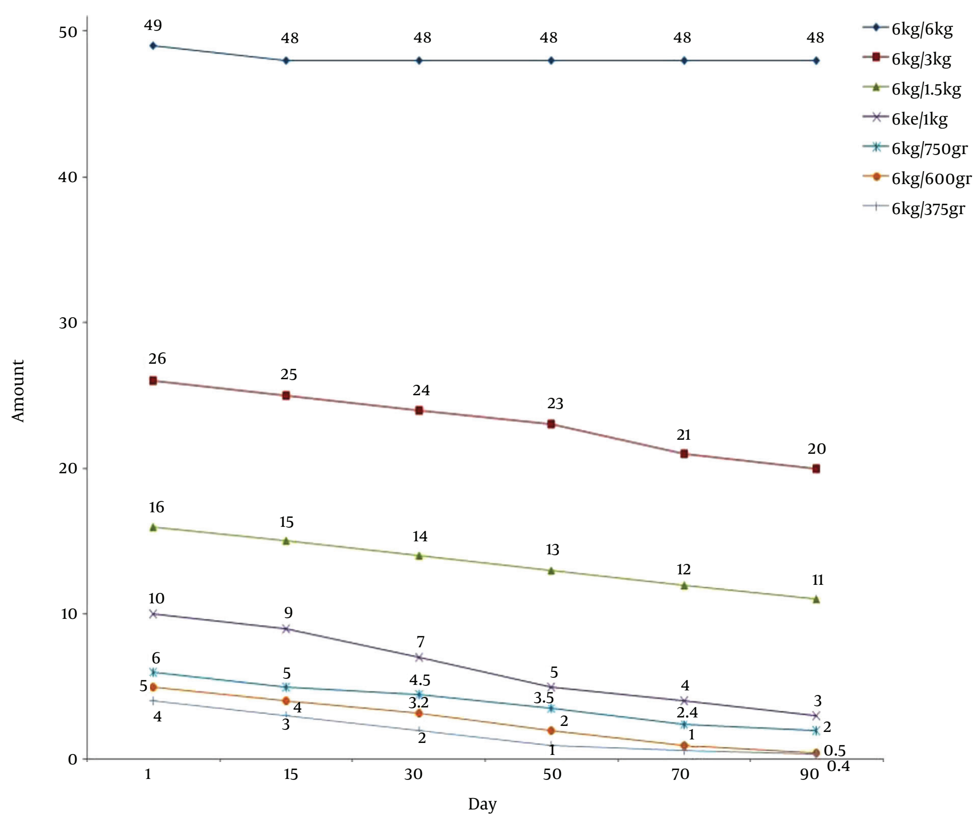

Figure 3. trend of Cr^{+6} concentration reduction for all vermicompost pilot of paint sludge and sewage sludge in mg/kg

compost pilot of paint sludge and biological sludge during decomposition of 90 days. As shown in the diagram, the amount of chromium decreased with increasing time in the vermicomposting process.

5. Discussion

Carbon and nitrogen are very effective in the decomposition of organic compounds in sewage sludge and paint sludge, and the ratio of carbon to nitrogen in vermicompost should be set to allow good decomposition. Total organic carbon is an important parameter for the metabolism of microbes. The microbes utilize the carbon

as the source of energy and some part of carbon was lost as CO_2 during the composting process. Carbon to nitrogen proportion is the most widely used parameter in composting. Generally, composting could be carried out under a wide range of initial C/N ratios, namely, 11 to 105, depending on the starting materials (5, 20, 24).

The optimal ratio of organic carbon to organic nitrogen, at the start of the vermicompost process, should be between 20 and 30 and the best ratio is 25.1 (21, 22). This ratio can be increased by mixing cellulosic materials, such as straw and garden debris, into waste mass, or be decreased by mixing the sewage sludge of sewage treatment plant; as shown in Figure 1, C/N ratio are initially very low and approximately between 5 and 7, thus the C/N ratio of all treatments was initially set at 25 by straw and garden debris. The variation of C/N at different intervals of vermicompost decomposition for three months is shown in Table 3. The decrease in the C/N ratio during the vermicompost process is indicative of more sewage activity and carbon use by microorganisms.

pH is a very important parameter, which influences the vermicomposting process (21). The variation in the pH of the system affects the microbial activity on the organic waste. According to previous studies, the optimal pH for *Eisenia fetida* has been reported to be from 6 to 9 (22). As shown in Figure 2, the pH of the sewage sludge used in this study was in the neutral range (6.8) while the pH of Saipa's paint sludge was in the alkaline range (6.8), which changed during the vermicompost process. The pH of the mixture of paint sludge and sewage sludge was initially poor in alkaline, and over time and the process of vermicompost, it gradually turned into a weak alkaline state and neutral in the decomposed samples. It should be noted that the microorganisms involved in the decomposition of organic matter, especially in aerobic conditions, tolerate the range of pH from 5.5 to 9.5 (28).

As far as pH changes during 90 days in pilots A, B, C, F, E, D, and G were concerned, the high pH of 8.6 at the beginning of the decomposition was due to the nature of the residual paint. However, over time and with the activity of the microorganisms, decomposition took place and CO_2 as the most important weak acidic agent was released as a result of aerobic decomposition reaction (even the possible anaerobic process in the lower parts of the mass), reducing the pH. Based on this, in pilots A and B, which had less sewage activity, pH changes were not significant. That is, in pilot B, pH was 8.4 on the first day, which dropped to 8.35 at the end of the first month, and after a period of 90 days, it reduced slightly to 8.2. However, in pilots F, E, D, and G, due to the presence of active bacteria, microorganisms, and worms in the organic matter, pH changes were higher compared with pilots A and B. In pilots F and G, pH fell from 7.7 to 7.3, and the reason for this difference was due to the presence and activity of bacteria, microorganisms, and worms. In other words, as the growth rate of earthworms and the activity of microorganisms in the sample increased, the volatile solids decreased, and a portion of the organic matter was converted to ammonia, which was removed after shuffling and mixing the vermicompost mass, hence slightly reducing the pH. Most studies have also confirmed these pH changes. Additionally, these pH changes can be attributed to sewage conversion of organic matter to interstitial compounds, such as organic acids in the lower parts of the mass, which is due to the reduction of oxygen and sometimes the "anaerobic" condition (25). Figure 2 shows that in samples F and G, where the activity of the worms was higher (the ratio of the sewage sludge to the paint sludge being "one to ten"), the pH reached neutral levels, while in samples A, B and C, it remained in the range between 7.9 and 8.5. In other words, the environment remained alkaline.

The organic matter decomposition in the mixture of sewage sludge and paint sludge in the pilots by microorganisms was accompanied with reduction of volatile solids. Microorganisms use carbon as a source of energy. As seen in Table 4, it can be stated that on the first day, the ratio of the organic volatile matter in the seven pilots A, B, C, F, E, D, and G was 60.6, 65, 72.8, 57, 78.3, 80.4, and 86.3, respectively, which after 90 days decreased to 53, 49, 42.4, 42, 40.7, 37, and 34.5, respectively.

Decomposition of biological and paint sludge organic matters, by microorganisms is simultaneous with volatile solid's reduction. Microorganisms use carbon as an energy source. As shown in Table 4, it is clear that on the first day, organic volatile compounds of samples in A, B, C, D, E, F, and G were about 60.6, 65, 72.8, 75, 78.3, 80.4, and 86.3m, which after 90 days, reduced to 53, 49, 42.4, 42, 40.7, 37, and 34.5. The maximum amount of volatile organic compound's reduction, was reached in F and G pilots (Table 4). Also, most heavy metals amount reduction, occurred in these pilots (samples with more biological activity). In three pilots with few mixing of biological sludge (A, B and C), organic matters amount decreased from 60.6, 65, 72.8 to 49, 53 and 42.4 because of limited biological decomposition in paint sludge by microorganisms. Results of decomposition of F and G is shown in Table 4. The reason for the reduction of organic compounds, was higher activity of bacteria, microorganisms, and worms in biological sludge of wastewater. Increasing the microorganism's growth and greater ability of worms, by using biological organic compounds, cause their reduction in masses. By analyzing the decomposition process, in 90 days, the significant reduced amount of organic solids in pilots, especially in F and G, could be explained (Table 4).

Figure 3 showed that *Eisenia fetida* could reduce lower amounts of chromium (F and G pilots), significantly. In total, it isn't clear how and by which mechanism, worms can undergo this procedure. During the mentioned process, some parts of chromium stays on worm's skin and body. In samples with high amounts of chromium (A, B and C), worms died and some parts of the chromium rebounded to bed. As it is shown in charts, the best removal of chromium, was in the mixture of 6 kg of biological sludge, plus 600 gr of paint sludge (this means that the biological sludge's amount was 10 times more than paint sludge). By storing heavy metals on skin and tissues of worms, after their death and decomposition in the body tissues, some amounts of absorbed metals rebounded to fertilizers bed and caused an increase in the amount of chromium in the study samples (25, 26). Figure 3 shows that chromium's amount, in 90 days and in best mixing ratio (10/1), decreased from 5 to 0.5. In other words, chromium elimination efficiency was 92%.

Eisenia fetida have to accumulate the heavy metals in their bodies from soil as well as different biological

Wastes during vermicomposting. The *Eisenia* and *Eudrilus eugeniae* are the most important species for effective reduction of metal toxicity from industrial solid waste (26, 27). Figure 3 shows that *Eisenia fetida* can reduce lower amounts of Cr^{+3}, significantly. Overall, it is not clear how and by which mechanism worms can do this procedure. During the mentioned process, some parts of heavy metals stay on worm's skin and body. In samples with high amounts of heavy metals, worms died and some parts of the heavy metals rebounded to bed. As indicated in the figures, the best removal amount of heavy metals, was with a mixture of 6 kg of sewage sludge, plus 600 gr paint sludge (this means that the sewage sludge's amount was 10 times more than paint sludge). By storing lead and zinc metals on skin and tissues of worms, after their death and decomposition of the body tissues, some amounts of absorbed metals rebounded to fertilizers bed and caused an increase in the amount of lead and zinc metals in the study samples. Figure 3 shows that Cr^{+3} amount in 90 days and in best mixing ratio (10/1) decreased from 5 to 0.2. The results showed that the optimum removal rate for heavy metals was found in the mixing ratio of 6 kg of sewage sludge with 600 g of paint sludge and in this ratio, the amount of Cr^{+6} decreased from 5 mg/kg to less than 0.2 mg/kg in 60 days. Regarding earlier studies, earthworms can decompose some pollutants, such as heavy metals, yet the amount of pollutants should not be too much. Earthworms in general (specially *E. fetida*) are highly resistant to many chemical contaminants, including heavy metals and organic pollutants in soil and have been reported to bio-accumulate them in their tissues. Researches on vermiculture has revealed that worms can feed upon a wide variety of materials from earth. Aristotle called them 'intestine of earth' (5, 23, 27). A study titled "Removal of aluminum, Lead and Nickle from industrial sludge via vermicomposting process" focused on feasibility of using vermicomposting by *Eisenia fetida* in treating electric industrial sludge. Electronic industrial sludge consists of metals, such as aluminum, nickel and lead. In this report, metal compounds elimination from electronic factory waste sludge, during the vermicompost process, was confirmed (8, 29) 42. One research, titled "Heavy metals remediation from urban wastes using three species using of earthworms (*Eudrilus eugeniae*, *Eisenia fetida* and Perionyx excavates)", showed that elimination of cadmium, lead, zinc and copper during the vermicompost process by using *Eudrilus eugeniae*, *Eisenia fetida* and Perionyx excavates in three types of wastes (MSW, MW and FW) in 60 days in *Eudrilus eugeniae* was more than other species. On the other hand, increasing the vehicles number, caused more industrial waste production, such as heavy metals. According to their health disorders, it is necessary to manage this hazardous compounds (30).

The heavy metals accumulated in earthworms tissues are analyzed in terms of concentration factors, such as the bioconcentration factors (BCFs) and the bioaccumulation factors (BAFs) with the assumption that the organisms have achieved chemical equilibrium with respect to a particular media or route of exposure (8, 30).

5.1. Conclusion

In this study, the amount of volatile solids and heavy metals reduction from Saipa automobile paint sludge was investigated by a vermicompost process mixed with different proportions of biological sewage sludge of the sewage treatment plant. The results of the analysis showed that the best removal rate of volatile solids and Cr^{+6} was with the mixing ratio of 6 kg of biological sludge with 600 g of paint sludge, which is a ratio of 10 to 1. Volatile solids in a sample with an optimal mixing ratio (ratio of 10 to 1) decreased from 80% to less than 37% mg in 60 days. Therefore the results showed that the optimum removal rate for heavy metals was found in the mixing ratio of 6 kg of sewage sludge with 600 g of paint sludge and in this ratio, the amount of Cr^{+6} decreased from 5 mg/kg to less than 0.2 mg/kg in 60 days. In the same ratio, the C/N ratio decreased from 27 on the first day to 13.5 on the 90th day. Also, the amount of volatile solids or organic solids in the mass was significantly reduced. Therefore, the *Eisenia fetida* worm, along with the microorganisms in the biological sludge, is capable of being active in high proportions of biological sludge mixed with paint sludge and has a very high ability to decompose organic matters, including volatile solids.

Acknowledgments

This paper was issued from Ph.D thesis of Rahman Ghomi Avili and financial support was provided by Science and Research Branch, Islamic Azad University, Tehran, Iran. (Grant Number E 870)

Footnotes

Authors' Contribution: Concept and design, Afshin takdastan and Gasemali Omrani; analysis of data, Rahman Ghomi Avili and Farideh Atabi; writing of the manuscript and revision, Afshin Takdastan and Rahman Ghomi.

Conflict of Interests: Authors declared no conflict of interest.

Funding / Support: The study was financially support by the department of Environmental Engineering at Science

and Research Branch, Islamic Azad University, Tehran, Iran.

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