

Research Article

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Assessment of content and chemical forms of arsenic, copper, lead, and chromium in sewage sludge compost as affected by various bulking agents

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Abstract: In current study, the effect of various organic substances as bulking agents (BAs) including wheat straw, pistachio hull wastes, and tree leaves at different levels (10, 25, 45% v/v) were investigated on total concentration and chemical forms of Cu, Pb, Cr, and As in sewage sludge (SS) compost prepared by windrow method. According to the results, the composting process (with/without BAs), due to losses of SS mass and volume, increased the total concentration of heavy metals (HMs) compared to the un-composted SS sample (RSS). Evaluation of HMs chemical forms in prepared compost sample without BAs application (CSS) showed that the composting process reduced the mobility factor of As (from 28% to 20%), Pb (from 11.6% to 9.3%), and Cr (from 14.5% to 9.2%) compared to the RSS. Application of three BAs considerably decreased the mobility factor of As (17.5–18.8%), Pb (4.8–7.9%), and Cr (1.4–6.8%) compared to CSS and RSS. Changes of Cu mobility in prepared compost samples showed an unclear trend, however in some treatments, due to transferred organic fraction into exchangeable and carbonate fractions, increasing of this factor was obvious. Generally, the composting appeared to reduce As, Pb, and Cr availability by stabilizing the three

metals and making them more stable and less mobile. In addition, the BAs application effect on HMs behavior of SS compost samples were so different and no specific kind of BAs can be recommended as a superior BAs in SS composting process.

Keywords: sewage sludge; compost; bulking agents; heavy metals; chemical fractionation

1 Introduction

In recent decades, the production of SS has increased with increasing the world's population, which no specific consumption is defined for them in wastewater treatment plants. Disposal or safe use of these materials requires a great deal of planning and effort in a way that does not harm public health or cause environmental problems (Kelessidis and Stasinakis, 2012). In some parts of world, SS has been used for many years to improve the conditions and fertility of agricultural lands, but its environmental aspects have not been given enough attention. The SS contains large amounts of organic matter and essential nutrients for plant growth, but existence of hazardous pathogens, toxic organic compounds and HMs in its composition, has caused that the application of this organic source to agricultural lands is associated with environmental risks (Smith, 2009). Therefore, in order to minimize the environmental impact of this source on soil, water and plant environments, SS should be properly stabilized before use.

The high costs and insufficient efficacy of different stabilization methods of SS such as heat treatment, heat drying, pasteurization, using beta and gamma ray radiation, aerobic and anaerobic digestion, alkaline stabilization, etc., as well as strict rules for prohibiting the discharge of SS into water and land resources has caused the alternative

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methods such as composting is suggested by researchers (Cieřlik et al., 2015). The biological composting processes of SS not only can stabilize and convert the organic matter to humus-like product as an excellent organic fertilizer, but also it could reduce the population of pathogenic agents (Alvarenga et al., 2015). On the contrary, the composting process adds structuring materials, increasing the final volume and diluting heavy metals (Alvarenga et al., 2015). To solve this problem, addition of some amendments such as lime and fly ash (Samaras et al., 2008) to SS was suggested in composting process, which could decrease the availability of HMs. The nature of these amendments is alkaline and their application may cause an increase in soil pH that limits its using in agricultural land, especially in alkaline calcareous soils of Iran. In addition, lime or other alkaline substances application could increase the ammonia emissions and consequently increase the environmental effects of ammonia release and decrease the nutrient status of the product. Thus, using various organic matters may be useful to reduce HMs availability via promoting humification process and also by dilution in the total mass (Doublet et al., 2011). In the other words, the presence of external organic compounds in composting process may have an effective role in converting the mobile parts of HMs into less mobile fractions (Doublet et al., 2011). In addition, due to low porosity and air conditions of SS, using these materials in SS composting process could also adjust sludge conditions (reducing sludge moisture content and adjusting the C/N ratio) as well as improve air condition in composting process (Yañez et al., 2009). Thus, these materials are also used as bulking agents (BAs).

Determination of HMs availability via the conventional extractors such as DTPA or EDTA, as single extraction methods, may not properly interpret the availability of each element, so it is necessary to determine the chemical forms of each HMs via sequential extraction methods. Determining the chemical forms of each element in composts can play an important role in identifying the risky behavior of HMs (Amir et al., 2005).

Until now, few attempts have been done to assess the effects of organic BAs on HMs concentrations of SS compost and their chemical forms. In current study, this hypothesis exists that the use of different organic BAs in composting process may reduce amount of HMs availability in SS compost. Therefore, the present study tries to study the effect of mixture of SS and three BAs including tree leaves, wheat straw, and pistachio hull wastes in the composting process on total concentration and chemical forms of As, Cu, Pb, and Cr in produced compost from SS.

2 Results and discussion

2.1 Chemical properties of produced SS composts

Investigating the chemical changes of the produced compost samples can be useful in interpreting the behavior of the studied HMs. Hence, the pH, EC, OC, and total N values of RSS and composted SS samples are shown in Figure 1. According to the results, in some cases

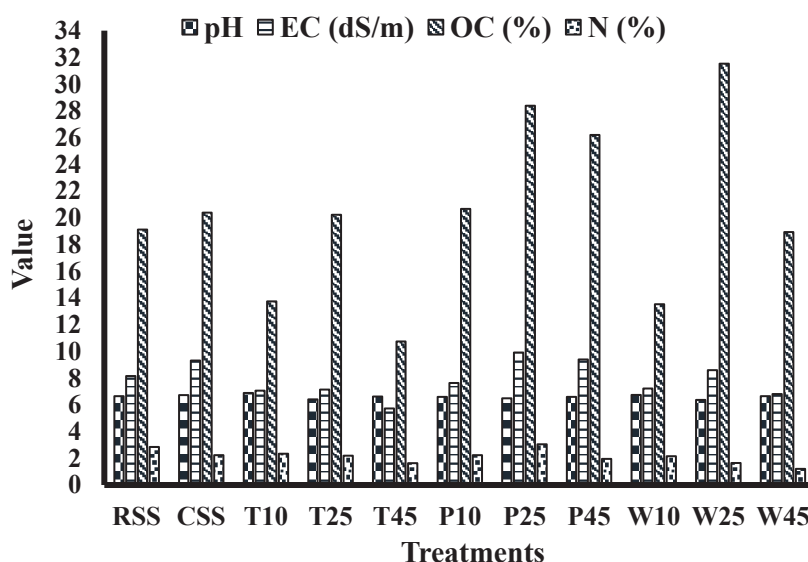


Figure 1: Variation of pH, EC, OC, and total N values in produced SS compost samples as affected by various BAs (Each abbreviation in x-axis is defined in Table 3).

of samples, the use of BAs reduced pH compared to the RSS and CSS. The highest and lowest amounts of pH were observed in T10 and W25, respectively. The pH values of produced compost samples were observed in the range of 6.3 to 6.9, which were in accordance with the acceptable level of the first class of compost (6 to 8) provided by the Iranian Institute of Standards and Industrial Research (ISIRI, 2011). In the present study, the application of composting process in some samples reduced the pH values, which may be due to the production of CO_2 and organic acids by microorganisms (Gunadi et al., 2002). On the other hand, there was an increasing trend in another samples. Jaafarzadeh Haghighifard et al. (2015) showed that the increase pH of MSS compost was due to the ammonia and mineralization of organic nitrogen (due to microbial activity) and the decrease pH was due to the nitrification and ammonium gas formation and release to the atmosphere and eventually release of H^+ ions (Wong et al., 2001a). It seems that differences in the type of ammonium and nitrate forms in the studied SS samples have caused pH changes, so that it can be said that the presence of more nitrate nitrogen in some samples decreased the pH and more organic nitrogen in another samples increased the pH. The difference in pH value in compost samples with different BAs can be due to the varying amounts of lignin and cellulose that are slowly degraded by the bacteria and thus effective in releasing ammonium and nitrate. According to the results of EC variation, the changes in EC values in the production composts have not followed a certain trend. Changes in EC values varied according to the types of BAs and their levels. High EC values in BAs of P (7.7 dS m^{-1}) compared to the other two BAs (2.6 and 0.6 dS m^{-1}) is an important reason for increased EC in produced compost with P. On the other hand, the decrease of EC values in other samples compared to the un-composted samples is the direct consequence of the increased concentration of nutrients, such as nitrate and nitrite (Bazrafshan et al., 2016), as well as the decreasing salt content per unit volume of compost due to addition of BAs. The highest and lowest values of this parameter were in samples P25 and T45, respectively. The decrease in the volume of MSS and the increase in soluble salts per unit volume of compost are the main reasons for the increase in EC values in compost samples without BAs. According to classification of compost quality (ISIRI, 2011), 6 samples of produced composts were in class I (less than 8 dS m^{-1}) and 4 samples of produced composts were in class II of compost (less than 14 dS m^{-1}). Grigatti et al. (2011) showed a decrease in EC of compost samples produced from MSS compared to non-composted samples. On the other hand, research by Tandy et al.

(2009) showed an increase in the EC of SS compared to non-composted samples due to the production of mineral compounds and a relative increase in the concentration of ions (decrease in mass of compost due to decomposition of organic matter and also raw materials for composting). As results of OC in samples, two BAs of P and W increased this parameter of compost and the BAs of T decreased this parameter of compost compared to RSS and CSS. It seems that lower values of the recalcitrant C fraction of organic matter (lignin and cellulose) of the BAs of T particle caused more decomposition of the compost OC and decreased this parameter in this treatment. As the results, the highest and lowest values of this parameter between produced composts were observed in samples of W25 and T45, respectively. According to the results, the OC values in the produced compost samples were in the range of 10.73 to 31.51%. Based on the acceptable level of OC (class I: more than 25% and class II: more than 15%) provided by ISIRI (2011), the obtained values of most samples have classified in class II of compost. Hernández et al. (2006) in the study of the production of sewage sludge compost with different BAs showed that the large amount of organic matter was reduced by the process of mineralization of many labile components by microorganisms. A study by Gao et al. (2015) showed that in the composting process, the amount of organic matter decreased due to mineralization and production of humic substances.

According to the results of total N in prepared samples, the composting process in most cases reduced this parameter compared to the non-composted samples, which is justified by the process of nitrate denitrification and sublimation of ammonium to ammonia over a high temperature ($58\text{--}63^\circ\text{C}$) during the composting process, however the trend of N changes in the treatments did not follow a specific order based on the kinds and levels of BAs. As results, the highest amount of this property was in the presence of P25. Based on the results, the N content in the produced compost samples were in the range of 1.21 to 3%, which based on the acceptable level of N content (class I: 1.25 to 1.66% and class II: 1 to 1.5%) of compost provided by ISIRI (2011), the values obtained are above the minimum value of 1.25% and classified in class 1 of compost.

2.2 Total concentration of HM_s in prepared SS compost samples

Table 5 shows the total concentration of some HMs in RSS sample and produced compost with/without BAs

application, after 12 weeks. A comparison of HMs total concentration in RSS with CSS (without BAs) shows that the composting process, due to losses of SS mass (biodegradation of organic matter), increased HMs total concentration in CSS. This finding is in agreement with previous studies (Cai et al., 2007; Zorpas et al., 2008). As seen in Table 5, application of each BAs in different volumes had different effects on HMs concentration in composted samples. For instance, application of 10% from each BAs caused an increase in total As concentration compared to CSS, but application of BAs in 45% (with exception of W45) reduced the total amount of As. This trend was also observed for Pb. It seems that in a low level of BAs, the preparation of suitable aeration conditions may improve the composting process and resulted in a decrease in SS mass and consequently increased HMs concentrations compared to the CSS. On the other hand, in high level application of BAs, HMs concentration in composted SS samples reduced due to dilution effect. The changes of total concentration of Cr and Cu as affected by BAs had unclear trend, however, generally can be shown the total concentration of Cr increased and Cu decreased. In evaluating the quality of compost produced from SS (from Isfahan wastewater treatment plant, Iran) as affected by different BAs (green plant waste (GW) and dry leaves and pruning waste (PW); ratio of 1:1, 1:2, and 1:3 $v_{(SS)}/v_{(Bas)}$), Nafez et al. (2015) showed that addition of BAs had a diluting effect on HMs and HMs amounts were lower in the 1:3 mixture than the 1:2 mixture. Their results also showed that BAs application decreased bioavailability of HMs, which were below the toxicity threshold for agricultural application. Malakootian et al.

(2014) investigated the possibility of producing compost from SS (Kerman wastewater treatment plant, Iran) and pistachio hull wastes as a BAs. Their results showed that addition of the Bas, due to dilution effect, decreased the total form of Pb and Cd in compost samples. As results of total forms of studied HMs, it can be concluded that the change of total concentration of HMs may not predict and interpret the behavior of each HMs in produced composts. Therefore, evaluation of chemical forms of each element could be useful to better understand the effect of BAs on behavior of HMs in composted SS.

2.3 Chemical forms of As

Since the process of composting has changed the total amount of HMs in each compost sample, thus assessment of relative percentage ([each form of element (mg kg^{-1})/total element (mg kg^{-1})] $\times 100$) variations of any chemical forms of elements can be better, for a more appropriate interpretation of chemical fractionation changes, than variations of chemical forms concentration. Chemical forms of As in RSS sample showed that (Figure 2) the highest form of As exist in the form of Fe-Mn-OX (43.6%), followed by Car (23.4%), Res (21.6%), OM (6.6%), and EX (4.6%) forms. Based on the results, the process of composting without BAs application increased the forms of Fe-Mn-OX (51.6%) and OM (9.6%), however reduced the forms of EX (2.8%), Car (17.23%), and Res (18.7%). Hanc et al. (2012) reported that composting process on kitchen and garden wastes increased the EX, reducible and oxidizable forms of As, while decreased the Res

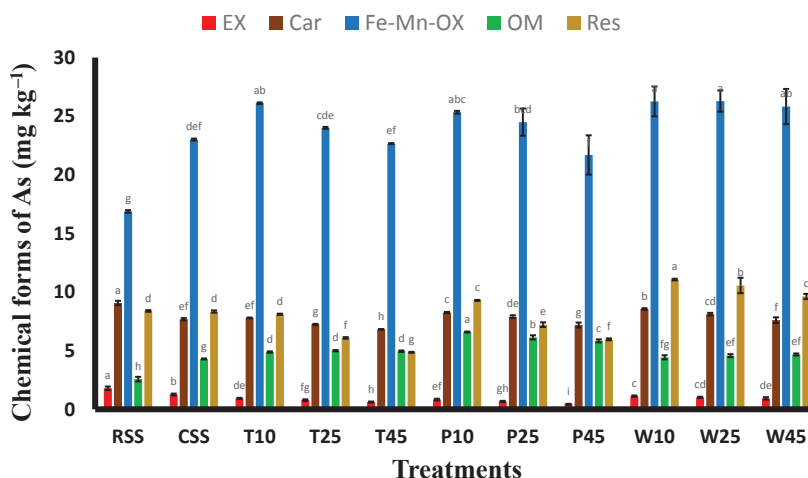


Figure 2: Chemical forms of As in RSS and produced compost samples. Different letters (in the same chemical forms of treatments) indicate significant differences among the mean of different treatments ($p < 0.05$). Error bars represent the standard deviation. (Each abbreviation in x-axis is defined in Table 3).

form. Application of each BAs in composting process has obviously made changes in the chemical form of As. Using of BAs in composting process increased the Fe-Mn-OX (50.4-56.8%) and OM (8.6-14.2%) forms; but reduced the EX (1-2.2%), Car (15.6-17.4%) and Res (12.1-21.5%) forms. Among the BAs, application of P45 and W10 had the highest and lowest effects on EX decreasing. The composting process has reduced the mobiles forms of As (EX and Car) in SS. According to the literature, chemical forms of EX, Car, Fe-Mn-OX, OM, and Res, have the highest to lowest availability of HMs, respectively (Saffari et al., 2019). The mobility factor (EX+Car/sum of chemical forms) of As in SS and compost samples are shown in Figure 3. As seen, the mobility factor for SS, CSS, and compost samples prepared by BAs were 28%, 20%, and 18-18.8%. In general, the results show that the use of BAs reduced the As mobility compared to the CSS and RSS samples. As exist in two forms of As_{III} (arsenite;

as dominate forms of As under oxic condition) and As_V (arsenate; as dominate forms of As under anoxic condition) in environment, which the toxicity and availability of As_{III} is much more than As_V (Jain and Ali, 2000). It seems that, under oxic conditions of composting process, arsenite oxidized to arsenate and this form of As adsorbed on Fe-Mn-OX (as reducible fraction) of compost (Newton et al., 2006), and ultimately the availability of As decreased followed the composting process. This finding is agreement with the study of Wang et al. (2018), which showed the ratio of arsenate to arsenite increased along with the composting process.

2.4 Chemical forms of Cu

Chemical fractionation of Cu (Figure 4) showed that the OM (41-61%) and Res (29-44%) forms were the dominant

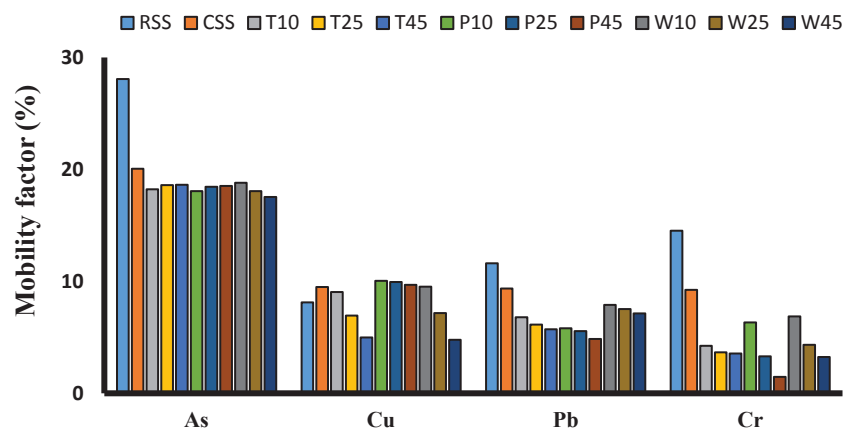


Figure 3: Mobility factor of As, Cu, Pb, and Cr in RSS and produced compost samples (Each abbreviation in legend is defined in Table 3).

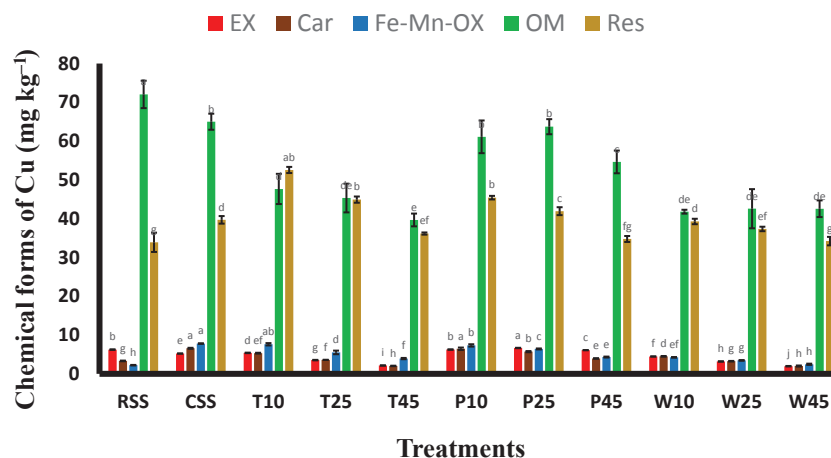


Figure 4: Chemical forms of Cu in RSS and produced compost samples. Different letters (in the same chemical forms of treatments) indicate significant differences among the mean of different treatments ($p < 0.05$). Error bars represent the standard deviation. (Each abbreviation in x-axis is defined in Table 3).

chemical forms of Cu in RSS and composted samples. In the contrary, the EX (2.3-5.8%), Car (5.6-16.5%), and Fe-Mn-OX (1.9-6.4%) forms of Cu exist in very low concentration in both RSS and composted samples. Previous studies (Fuentes et al., 2004; Negreanu-Pirjol et al., 2012) showed that Cu tends to form very stable complexes with organic ligands (bound to two or more organic functional groups mainly carboxylic, carbonyl and phenolic), thus, identifying the OM form as the dominant form of Cu in SS and their composts was predictable. This form of Cu is recognized as an immobile fraction that could only be available under extreme oxidizing conditions. Composting of SS (without BAs) reduced OM (52.3%) and EX (4.2%) forms, but increased the forms of Res (32%), Fe-Mn-OX (6.3%), and Car (5.3%). In agreement with our finding, Qiao and Ho (1997) showed that the composting process reduced the OM form of Cu and transferred it into Car, Fe-Mn-OX and EX fractions. Miaomiao et al. (2009) assessed the effect of composting process on some HMs forms in SS and showed that composting process increased and decreased the readily extractable fractions (EX and Car) and Fe-Mn-OX of Cu in SS, respectively. Addition of BAs in composting process of SS increase the Res (33.4-44.3%) form and decrease OM (40.2-52.6%) form in comparison with CSS. The decomposition of the organic matters (as a result of the composting process) results in the release of the Cu complex with the organic component and reduced the OM form. Contrary to the findings of this research, Cai et al. (2007) reported that composting process of SS increased OM form and converted sulfide form into the OM form. Based on the mobility results of Cu

(Figure 3), the CSS had a mobility factor about 8%, which composting process without BAs increased it by 9.5%. The mobility factor of Cu in composted samples with BAs have been affected by BAs type and its application level. For instant, application of T45 and W45 lead to reduce the mobility factor by 4.7% and 5%, respectively. But in P10 and W10 treatments, mobility factor increased by 10% and 9.51%, respectively. Different kinds of BAs appear to act as a HMs adsorbent and by forming an organic complex and enhancing the organic form of this element, reduce the EX of Cu.

2.5 Chemical forms of Pb

Investigating the chemical forms of Pb in RSS indicates the superiority of the Res form (39.7%) as a dominant form of Pb (Figure 5), which is reported by others (Qiao and Ho, 1997; Shrivastava and Banerjee, 2004). After the Res form, the Fe-Mn-OX (26.1%), OM (22.5%), Car (6.4%) and EX (5.1%) forms had the highest to lowest fractions of Pb in RSS sample. The composting process has led to the appearance of the two forms of Res and OM, as the dominant forms of Pb. Composting process (without BAs) increased the Res (56.1%) form and reduced other forms compared to RSS sample. The application of BAs in the composting process compared to CSS reduced the forms of EX (2.5-3.9%), Car (2.3-4%), and Res (51.7-55.57%), but increased other forms. Compared to the fractionation results of As and Cu, the use of all BAs in each levels can reduce the Pb mobile forms compared to CSS (Figure 5).

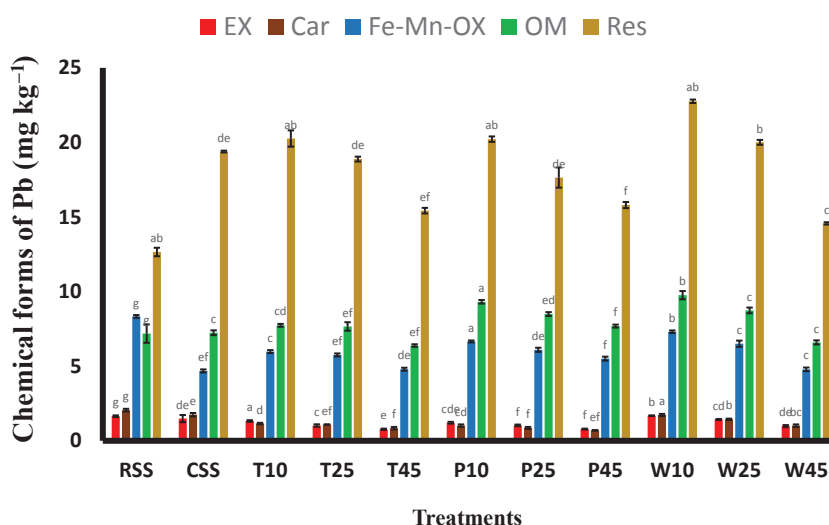


Figure 5: Chemical forms of Pb in RSS and produced compost samples. Different letters (in the same chemical forms of treatments) indicate significant differences among the mean of different treatments ($p < 0.05$). Error bars represent the standard deviation. (Each abbreviation in x-axis is defined in Table 3).

The Pb mobility factor in SS was 11.6% that composting processes without BAs reduced the mobility factor to 9.3%. The use of BAs significantly reduced the mobility factor in comparison with the CSS and RSS. The lowest mobility factor (4.9%) was observed in treated sample with P45. Decreasing in Pb mobility in compost samples may be related to increasing dissolved organic carbon (DOC) following the compost process (Qiao and Ho, 1997). Pb can facilitate the removal of DOC from SS by adsorption of positively charged Pb-organic complexes and cation bridge, which consequently decreased its availability (Singh and Kalamdhad, 2013).

2.6 Chemical forms of Cr

The results obtained from the chemical forms of Cr in RSS show that OM (70.4%), Res (13%), EX (9.9%), Car (4.6%), and Fe-Mn-OX (9.1%) forms are the most to the least fractions of Cr fractionation, respectively (Figure 6). Numerous studies have shown that Cr is often present in the oxidizable and Res fractions, which is the reason why it has low availability (Tytła, 2019). The research of Wong et al. (2001b) and Walter et al. (2006) showed that more than 90% and 95% Cr of sewage sludge are in the OM and Res fractions. According to the results, in the end of composting process (without BAs application), OM, EX, Fe-Mn-OX, and Car forms decreased and Res form increased, compared to the RSS. Hanc et al. (2012) showed that composting process on kitchen and garden wastes decreased the EX and reducible forms of Cr. They stated that “reducing in reducible forms of Cr may

be caused by reduction of $\text{Cr}_{(\text{VI})}$ to $\text{Cr}_{(\text{III})}$ by microorganisms, followed by the combination of $\text{Cr}(\text{III})$ with decomposed organic materials”. The application of BAs in the process of composting reduced the EX (0.9-3.8%), Car (0.5-3.3%), Fe-Mn-OX (0.6-1.7%) forms and increased Res (39-60.2%) form in comparison with CSS. The mobility factor of Cr (Figure 3) in produced compost samples shows that composting process (without BAs) decreased this factor from 14.5 (for RSS) to 9.2% (for CSS). The use of BAs had a significant decrease in Cr mobility factor than RSS, which the highest (1.46%) and lowest (6.3%) decrease were observed in P45 and P10, respectively. Zheng et al. (2007) in a study of the stabilization of Cr and Ni in the composting process showed that the EX, Car, Fe-Mn-OX, and OM forms were decreased, while the Res form was increased.

3 Conclusions

According to the results of this study, despite the increase in total concentration of the studied elements, composting process decreased the availability of these elements. Adsorption of As on Fe-Mn-OX chemical form in result of the oxidation of arsenite to arsenate, assisting Pb in the removal of produced DOC in the composting process, and converting $\text{Cr}_{(\text{VI})}$ to $\text{Cr}_{(\text{III})}$ by composting microorganisms and combining this form of Cr with decomposition organic materials in compost, are introduced as the main factors in reducing the mobility of As, Pb and Cr in composting process. In addition, Cu, unlike the other studied elements, has a strong tendency

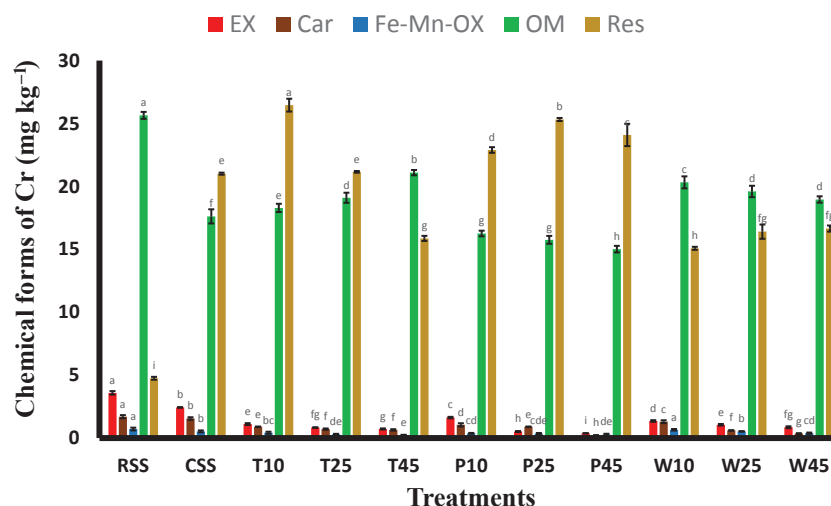


Figure 6: Chemical forms of Cr in RSS and produced compost samples. Different letters (in the same chemical forms of treatments) indicate significant differences among the mean of different treatments ($p < 0.05$). Error bars represent the standard deviation. (Each abbreviation in x-axis is defined in Table 3).

to adsorb organic forms and due to the composting and decomposing process, organic forms was converted to the mobile fractions and increased mobility of Cu in some treatments. Application of BAs increased the stability and decreased the mobility factor of the studied metals compared to produced compost without BAs. Generally, it can be shown the use of composting process in the stabilization of SS with BAs application can reduce the availability of HMs more effectively than composting of SS without treatment.

Experimental

Chemicals

The chemicals used in this study, including magnesium chloride (MgCl_2), sodium acetate (NaOAc), hydroxylamine hydrochloride ($\text{NH}_2\text{OH}\cdot\text{HCl}$), acetic acid (CH_3COOH), Nitric acid 65% (HNO_3), hydrogen peroxide (H_2O_2), ammonium acetate ($\text{CH}_3\text{COONH}_4$), sulphuric acid (H_2SO_4), potassium sulfate (K_2SO_4), potassium hydroxide (KOH), sodium hydroxide (NaOH), ammonium heptamolybdate tetrahydrate ($(\text{NH}_4)_6\text{Mo}_7\text{O}_{24}\cdot 4\text{H}_2\text{O}$), potassium antimony tartrate ($\text{C}_8\text{H}_8\text{KO}_{12}\text{Sb}$), ascorbic acid ($\text{C}_6\text{H}_8\text{O}_6$), acetone ($\text{C}_3\text{H}_6\text{O}$), sodium hydrogen sulfite (NaHSO_3), ammonium persulfate ($(\text{NH}_4)_2\text{S}_2\text{O}_8$), potassium dihydrogen phosphate

(KH_2PO_4), and barium acetate ($\text{Ba}(\text{OAc})_2$) were purchased from Merck (Darmstadt, Germany).

Preparation and characterization of SS and BAs

In order to product SS compost samples, the dried SS (raw SS: RSS) sample in sludge-drying lagoons was collected from the Kerman wastewater treatment plant, Kerman, Iran. Different kinds of bulking materials including tree leaves of broad-leaved tree species, wheat straw, and pistachio hull wastes prepared from Khanok area, Kerman, Iran, were collected for SS composting processes. Some chemical properties of dried SS sample and BAs are shown in Tables 1 and 2, respectively, which were measured by routine methods (Brunner, 1978). In a brief, the pH and the electrical conductivity (EC) of the SS and BAs were measured in a suspension of 1:5 materials and distilled water. Organic carbon (OC), cation exchange capacity (CEC), total nitrogen (N), total phosphorus (P), and total potassium (K) were measured at 550°C for 2 h in a muffle furnace, by replacing cations with barium acetate (measured by flame photometer (Jenway PFP7)), by Kjeldahl method (reduced to ammonia in a concentrated sulfuric acid solution), by yellow colorimetric method (vanadium phosphomolybdate method, measured by flame spectrophotometric technique (Cary 50 UV-VIS spectrophotometer)),

Table 1: Some chemical properties of sewage sludge in this study.

Property	Value	Property	Value	Property	Value
pH	6.6	K (%)	1.51	Available-As (mg kg^{-1})	16.31
EC (dS m^{-1})	8.13	CEC (cmol kg^{-1})	35.6	Available-Pb (mg kg^{-1})	0.80
OC (%)	19.28	Total-As (mg kg^{-1})	38.70	Available-Cr (mg kg^{-1})	0.83
N (%)	2.83	Total-Pb (mg kg^{-1})	31.20	Available-Cu (mg kg^{-1})	7.23
C/N (%)	6.81	Total-Cr (mg kg^{-1})	37.36		
P (%)	1.34	Total-Cu (mg kg^{-1})	118.30		

Table 2: Chemical properties of bulking agents used in current study.

Bulking agents	pH	EC (dS m^{-1})	OC	N	C/N	As	Pb	Cr	Cu
			(%)						
Tree leaves	7.5	0.6	46.78	0.69	67.80	ND*	ND	ND	7.6
Wheat straw	6.7	2.6	51.74	0.85	60.87	ND	ND	ND	11.4
Pistachio hull waste	7.7	7.7	48.56	2.8	17.34	ND	ND	ND	21.3

ND – Not detected

by aqua regia (HNO_3 and HClO_4) technique (measured by flame photometry (flame photometer: Jenway PFP7)), respectively.

method (Thompson et al., 2001) and transferred to the laboratory to measure the concentration of total As, Pb, Cr, and Cu, as well their chemical forms.

Production of SS compost samples

In this study, the windrow method was used to prepare SS compost samples. For this purpose, SS samples were mixed with different ratios of BAs [0%, 10%, 250%, and 45%, v/v] separately (Table 3). Each compost pile was built with a length of 2 m, width of 1 m, and height of 1.5 m. The water content (moisture 40-50%) and temperature was controlled during the composting process. Compost aeration was carried out every 5 days, which continued until the end of the composting process. After 12 weeks, the mature compost samples were collected (3 samples in each treatment) via TMECC¹

Total and chemical fractions of HMs

For extraction of HMs total concentration in compost samples, 1 g of each compost was digested by 10 mL of concentrated 4 M HNO_3 at 80°C for overnight in an oven (Sposito et al., 1982) and after filtering with filter paper, the concentrations of As, Pb, Cr, and Cu were measured by flame atomic absorption spectroscopy (Varian Spectra AA-10). Chemical forms of each HMs were determined using sequential extraction method of Tessier et al. (1979). Outline of the method is presented in Table 4. After each successive extraction, the supernatant was centrifuged

Table 3: Experimental design of SS compost production.

CSS	Composted SS sample without BAs addition	P25	75% (V) SS + 25% (V) pistachio hull waste
T10	90% (V) SS + 10% (V) tree leaves	P45	55% (V) SS + 45% (V) pistachio hull waste
T25	75% (V) SS + 25% (V) tree leaves	W10	90% (V) SS + 10% (V) wheat straw
T45	55% (V) SS + 45% (V) tree leaves	W25	75% (V) SS + 25% (V) wheat straw
P10	90% (V) SS + 10% (V) pistachio hull waste	W45	55% (V) SS + 45% (V) wheat straw

C – compost, SS – sewage sludge, T – tree leaves, P – pistachio hull wastes, W – wheat straw

Table 4: Summary of the sequential extraction procedure used in this study.

g compost : mL solution	Extracting solution	Shaking time (h)	Chemical form of HMs	Symbol
1 : 8	1 M MgCl_2 (pH = 7)	1 h in 25°C	Exchangeable	EX
1 : 8	1 M NaOAc (pH = 5 CH_3COOH)	5 h in 25°C	Carbonate-bound	Car
1 : 20	0.04 M $\text{NH}_2\text{OH}\cdot\text{HCl}$ in 25% CH_3COOH (pH = 2 HNO_3)	6 h in water bath 95°C	Bound to Fe/Mn oxyhydroxides	Fe-Mn-OX
1 : 8	30% H_2O_2 + 0.02 M HNO_3	2 h in water bath 85°C	Organically-bound	OM
	30% H_2O_2 + HNO_3	3 h in water bath 85°C		
	3.2 M $\text{CH}_3\text{COONH}_4$ + 20% HNO_3	0.5 h in 25°C	Residual	Res
1 : 8	4 M HNO_3 + 36% HCl	0.5 h in water bath 95°C		

Table 5: Total concentration (mg kg^{-1}) of HMs in RSS and composted SS samples.

	RSS*	CSS	T10	T25	T45	P10	P25	P45	W10	W25	W45
As	38.7	44.6	47.8	43.3	39.9	50.4	46.2	41.2	51.7	50.5	48.4
Pb	31.5	34.2	36.9	33.6	28.4	38.6	34.6	29.8	42.9	38.9	27.9
Cr	36.8	44.6	46.1	41.3	38.5	42.8	41.5	40.8	39.1	38.5	36.7
Cu	119.2	127.1	118.9	103.1	82.8	126.8	124.5	104.1	95.0	90.7	82.1

* Each abbreviation is defined in Table 3

(centrifugation at 8500 RPM for 15 min), filtered, and measured by flame atomic absorption spectroscopy.

Statistical analysis

The Duncan's multiple-range test procedure and other statistical analyses were calculated by Microsoft Excel 2010, SAS 9.1.3 and SPSS V19.

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