Poll Res. 40 (May Suppl. Issue) : S37-S44 (2021)
Copyright © EM International
ISSN 0257–8050

CHARACTERISTICS OF LEAD-SOIL CONTAMINATION IN CINANGKA, BOGOR INDONESIA AND DESIGNING SUITABLE BIOCHAR FOR ITS REMEDIATION

HIMAWAN[1,2], PRABANG SETYONO[1], SUNTORO[1] AND SENTOT BUDI RAHARDJO[1]

[1] *Postgraduate Program of Sebelas Maret University, Surakarta Indonesia*
[2] *Department of Chemistry Cenderawasih University, Jayapura Indonesia*

(Received 4 December, 2020; accepted 5 January, 2021)

ABSTRACT

Lead pollution of soil is a serious global problem due to risk of entering food chain and groundwater contamination. Anthropogenic activities such as mining, smelting, agriculture, and industry have played an important source of Pb in soil. Heavy metal is non-biodegradable and persists for long time in natural environment. Cinangka Village is a Pb-polluted area impacted from the processing of used lead-batteries since 1978. Although serious efforts of international and Indonesian government had been conducted in 2014, the remediation has not yet been completed because of insufficient resources. Therefore, it needs periodical monitoring and sustainable remediation. Biochar is a potential ubiquitous low-cost material for heavy metal-polluted soil remediation whose properties depend on its production conditions. This research aimed to study the condition of lead pollution in soil and groundwater in the former battery recycling and residential areas in Cinangka Bogor. Specifically, the study was focused on Pb profile on the soil horizon top soil, Pb groundwater level, its environmental risk assessment, and designing suitable biochar for lead immobilization. The total, fractions concentration, and leachability of Pb phytotoxicity characteristic leaching procedure (TCLP) were performed with AAS. The results showed that the soil in the former recycling center was acidic and contained Pb above 11,000 ppm which was dominated by reducible fraction (F2) and acid-soluble fraction (F1) having extreme contamination degree and very high environmental risk. Vertically until 45 cm depth, the deeper the soil layer the lower the Pb content. On the contrary, soil and groundwater in residential areas did not show any lead contamination. Biochar derived from mineral-rich material pyrolyzed at lower temperature is recommended to remediate the contaminated site.

KEY WORDS : Cinangka, Lead, Soil contamination, Ecological risk, Biochar design.

INTRODUCTION

Pollution occurs when an element or substance is introduced into the environment in a concentration higher than its natural background as a result of human activities and causes a net negative impact on the environment (Imeri *et al*., 2019). Toxic metals contamination has increased steadily in industrial era. Lead is a toxic heavy metal widely used in industry and is considered as a persistent pollutant in soil whose half-life of thousands of years (Orellana *et al*., 2019). Lead-contaminated soil has attracted many researchers to study the effect and solve the problem (Rizwan *et al*., 2016; Puga *et al*., 2015).

Legacy of Lead contaminated soil in Cinangka is a serious problem due to the health impact and risk of entering the food chain and groundwater contamination. The clean-up process of *in-situ landfill* encapsulation has been conducted from 2011-2014 covering 2,850 cubic meter waste of 15,726 tons of hazardous material collected from 4-hectare soil targets. Although succeeds, the project does not solve entire problem. Contaminated soil replacement following the clean-up process in 2014 only covers a small portion of the target land so there is still potential for agricultural land contamination.

As a semi-agrarian village, Cinangka, inhabited by 14,228 people, relies on the use of agricultural land so the level and ecological risk due to lead-pollution needs to be evaluated to ensure food security. Pb concentration in the leachate of TCLP test should be below the nonhazardous regulatory limit of 5 ppm (Huang *et al.,* 2016). Although Indonesia does not have soil quality standards, the government has stated Government Regulation No 101-2014 about the management of hazardous and toxic waste (HTM / B3). Anthropogenic activities such as mining, smelting, agriculture, and industry have played an important source of Pb in soil. Heavy metal is non-biodegradable and possesses long-term persistence in soils (Mahar *et al.,* 2015), therefore effective remediation measures are needed (Khalid *et al.,* 2016).

Conventional methods of contaminated soil remediation such as excavation, soil washing, and landfilling are costly and not compatible with farmland soil in developing countries (Khalid *et al.,* 2016). The more sustainable method having lower cost, improving soil quality, and adding economic benefits is being sought (O'Connor *et al.,* 2017). Immobilization is considered to be effective *in situ* best available method in the remediation of lead-contaminated agricultural soil in developing countries. Immobilization (chemical fixation) is focused to diminish lead mobility in contaminated soil to prevent plant uptake and groundwater contamination. Chemical stabilization/ immobilization of lead (metals) with low-cost amendments is another side of soil remediation. Biochar, a carbon-rich porous material produced from pyrolysis of agriculture byproduct, has widely applied due to its alkaline and good sorptive properties for metals and organics contaminant (Puga *et al.,* 2015; Ahmad *et al.,* 2016). This research not only thoroughly explore lead content and its environmental risks but also over a strategy of preparing biochar for effective, low-cost soil remediation.

Sequential extraction procedure (SEP) is a widely accepted method in assessing bioavailability and environmental risk of metal contamination in the soil. This procedure enables a better ecological assessment than total metal concentration. A simple, three steps, SEP method proposed by the Community Bureau of Reference (BCR) were performed in this study (Nemati *et al.,* 2011; Lu *et al.,* 2017). In SEP, metal concentration in the most mobile fraction is distinguished from other more stable fractions. In BCR procedure, metal in soils is defined operationally into four geochemical fractions/phases i.e, acid-extractable (F1), bound to Fe/Mn oxides (F2), bound to organic matter (F3), and the residual fraction (F4). Metal concentration in F1 is the most mobile fraction and determines the level of environmental risk of the metal (Nemati *et al.,* 2011; Zhang *et al.,* 2015; Yang *et al.,* 2018; Tytla, 2019).

Biochar is potentially used as an effective and low-cost amendment. Several important properties of biochar were pore size and volume, specific surface area, pH, ash or mineral content, and functional groups. These properties are influenced by types of organic matter feedstock, pyrolysis condition, and post-treatments. The choice of raw material, temperature and duration of pyrolysis, as well as biochar pore cleansing determine its performance in metal immobilization. The main goals of this study are (i) Explore the profile of contaminant (lead) level in top soil and groundwater (ii) Identify Pb-geochemical fraction and assess ecological risk of soil contamination (based on total concentration, TCLP, and SEP) (iii) designing the best biochar needed to remediate the soil.

MATERIALS AND METHODS

Contaminated soil was taken from Cinangka village in Bogor Regency, West Java, Indonesiaat coordinate of 6°35'28.98'' S and 106°41'34.27'' E as showed in Figure 1. Majority (72.3% of 4,061) households in this village use surface water. Among 14,288 people in Cinangka, 2,160 people are farmers and farm laborers. People who potentially depended on agricultural land are about 45.3%.

Research work was conducted in November 2019. Soil samples (at 0-15, 15-30, and 30-45 cm depth) from five locations in ex-smelter and residential areas were taken, digested in aqua regia, and measured with atomic absorption spectrophotometer (AAS) Shimadzu AA 6500 (Wiater, 2019; Mahar *et al.,* 2015). Electric conductivity (EC) and soil pH (H_2O) were measured in a 1:2.5 soil-water ratio. Composite soil samples from ex-smelter and residential areas were studied with x-ray fluorescence spectroscopy (XRF Shimadzu) to determine elemental composition. Water sample taken from five locations at residential area, was acidified with nitric acids, and concentrated prior to AAS measurement.

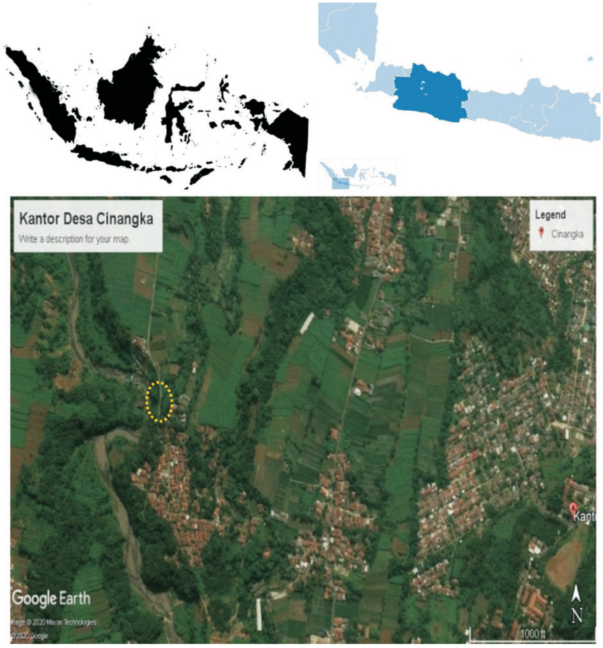

Fig. 1. Satellite image of sampling location

The SEP method of modified BCR (Nemati *et al.*, 2011; Lu *et al.*, 2017) was carried out to fractionatePb in soil into the geochemical phases. The fractions were acids-extractable (F1), bound to Fe/Mn oxides (F2), bound to organic matter (F3), and the residual fraction (F4). The three solutions used were: 0.11 M acetic acid; 0.5 M $NH_2OH.HCl$ pH 1.5; and twice of 8.8 M H_2O_2 then 1 M NH_4OAc at pH 2. Residual fraction remaining in the last step was extracted with aqua regia (Lim *et al.*, 2013; Rodriguez *et al.*, 2015; Mahar *et al.*, 2015). After centrifugation at 2500 rpm for 20 min and separation, the solid was then washed with 20 mL deionized water, shaken for 15 min, centrifuged at 2500 rpm for 20 min and filtered. The leadcontent in each fraction was quantified with AAS.

To assess the ecological impact of contaminated soil, the TCLP and some indices were employed. Values of geoaccumulation index (I_{geo}), contamination factor (CF), monomial potential ecological risk, E_r and Risk Assessment Code (RAC) were used in this research (Aktaruzzaman *et al.*, 2014; Tytla, 2019). TheI_{geo} compares current concentration with pre-industrial level logarithmically. The CF divides current level with background value based on world surface rock average given by Martin and Meybeck, (1979). Description of indices employed in this research is summarized in Table 1. The characteristics of contaminated soil (texture, pH, mineral content, Pb-level and share between geochemical fraction) are evaluated to design the suitable biochar for remediation based on literature review.

RESULTS AND DISCUSSION

Profile of contaminant (lead) level in top soil.

Sample soils from different depth is analyzed for Pb content, EC, and pH. The analysis of elemental composition (XRF), texture, and organic content are performed to topsoil in ex smelter area. Condition of ex-smelter and residential areas are summarized in Table 2 and 3.

Based on particle size and loss on ignition analysis (LOI), the soil is classified as loam having 18.9% clay particle and low organic substance. The average of Pb content in surface soil (0-15 cm) around the former smelter furnace is 11,192 ppm (between 10,051 and 11,754 ppm) and decreases in the underlying soil layers (Table 2). The upper soil layer is important for most annual plants and this surface-level exceeds 27 times of the WHO upper limit of 400 ppm. The level also exceeds 31 times of Chinese and 37 times of Australian standards (Yang *et al.*, 2018). In this soil layer also found remnants of battery processing such as lead slag and some battery component debris (Figure 2). Although this level is far below the highest level of soil ever reported (200,000 ppm) (Blacksmith Institute, 2014), soils with this level are still classified as hazardous and toxic material (HTM/B3). In the deepest

Fig. 2. Hazardous and toxic material (HTM) found at the location of ex-smelter.a: membrane, b: casing, c: battery separator, d: Pbslag.

Table 1. Pollution Indices and Ecological Risk criteria

	Geoaccumulation Index, I_{geo}	
Equation	Category	Description
$\mathbf{I_{geo} = {}^2\log [C_n/1.5\ B_n]}$	$I_{geo} \le 0$	Unpolluted
C_n : metal concentration	$0 < I_{geo} \le 1$	unpolluted to moderately polluted
B_n : metal level in pre-industry	$1 < I_{geo} \le 2$	moderately polluted moderately to
	$2 < I_{geo} \le 3$	heavily polluted heavily polluted
	$3 < I_{geo} \le 4$	heavily to extremely polluted extremely
	$4 < I_{geo} \le 5$	polluted
	> 5	
	Contamination factor, CF	
Equation	Category	Description
$\mathbf{CF = C_m / C_b}$	$CF < 1$	Low contamination
C_m : metal measured concentration	$1 \le CF < 3$	moderate contamination
C_b : metal background concentration	$3 \le CF < 6$	considerable contamination
	$CF > 6$	very high contamination
	Potential Ecological Risk Factor, E_r	
Equation	Category	Description
$\mathbf{E_r = T_f C_n / C_o}$	$E_r < 40$	Low risk
T_f : toxic response factor	$40 \le E_r < 80$	Moderate risk
C_n : metal concentration	$80 \le E_r < 160$	Considerable risk
C_o : metal reference value	$160 \le E_r < 320$	High risk
	$320 \le E_r$	Very high risk
	Risk Assessment Code, RAC	
Equation	Category	Description
	%RAC < 1	No risk
RAC = [F1+F2]/[F1+F2+F3+F4]	1 < %RAC < 10	Low risk
	10 < %RAC < 30	Moderate risk
Fn : metal concentration in fraction n	30 < %RAC < 50%	High risk
of BCR	RAC > 50	Very high risk

Table 2. Soil properties in ex-smelter soil horizon

Soil	A		B		C		D		E		Average	
depth	pH	EC	pH	EC	pH	EC	pH	EC	pH	EC	pH	EC
00-15	5,11	0,13	4,73	0,19	5,33	0,18	4,94	0,16	5,47	0,14	**5.12**	**0.16**
15-30	4,76	0,16	4,51	0,13	4,88	0,17	4,81	0,19	5,01	0,14	**4.79**	**0.158**
30-45	4,70	0,16	4,49	0,13	4,63	0,20	4,86	0,20	4,76	0,16	**4.69**	**0.17**

Table 3. Condition of topsoil (0-15 cm) and groundwater in residential areas

Sample	Soil O_1	Soil O_2	Soil O_3	Soil O_4	Soil O_5	Average
Pb (ppm)	63.7	68.1	60.7	55.0	71.4	**63.8**
pH	5.89	6.1	6.1	5.9	6.4	**6.1**
EC mS/cm	0.10	0.09	0.11	0.12	0.08	**0.10**
Sample	Water1	Water2	Water3	Water4	Water5	Average
Pb (ppb)	BDL	37	BDL	BDL	28	**32.5**
pH	7.0	6,7	6.8	6.4	6.6	**6.7**
EC mS/cm	< 0.01	0.01	< 0.01	0.01	< 0.01	**< 0.01**

studied soil layers (30-45 cm), Pb level was significantly decreased to 466 ppm or less than 1/24 of surface content. This fact will reduce the risk of lead movement into ground-water. Profile of Pb level in contaminated soil layers is depicted in Figure 3.

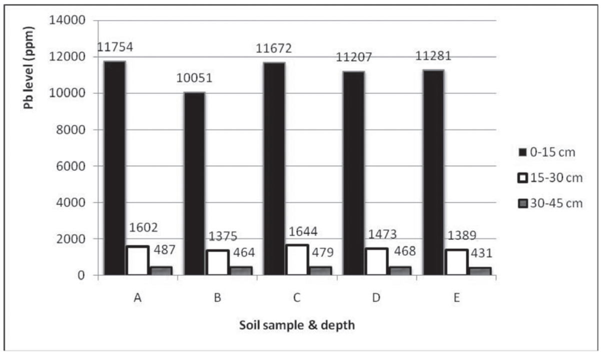

Fig. 3. Profile of Pb content in each soil depth at ex-smelter area

Table 2 also shows that all soil samples have acidic properties (pH ±5) below the quality standard (6.5-8.5). The soil has probably experienced pollution due to disposal of battery acid (sulfuric acid). In deeper layers of the soil, unlike Pb level that decreases significantly, the pH value of the soil tends to be fixed or only slightly changed. This acid needs to be neutralized to improve soil quality and mitigate risk of metal mobility.

There is no high Pb level found on the land around the residential areas (Table 3). Soil in this area contains much lower Pb, has lower electric conductivity and pH value. The average of Pb level was 63.8 ppm and there are no Pb levels that exceeded the WHO limit. Most water samples taken around settlements area have Pb levels below the measurement threshold. Two measurable samples (after being concentrated 5 times) have Pb levels of 28 and 37 ppb. This means that well waters in residential areas meet health requirements below 50 ppb. This result is slightly different from the results of Adryansyah *et al.* (2019) which showed the levels of Pb in water 58 - 123 ppb. Differences in sampling time and locations may be the cause of these differences.

Degree of contamination and ecological risks

To assess the ecological impact of contaminated soil comprehensively, the TCLP and some indices were employed. The result of TCLP test and SEP were shown in Table 4, whereas the values of pollution indices and ecological risk were presented in Table 5.

Based on Indonesia Government Regulation Number 101-2014, the TCLP-A value and total concentration (TK-A) for Pb are 3 and 6,000 ppm. This means that ex-smelter soils having a TCLP value of 56.2 ppm and TK-A of 11,192 ppm are categorized as hazardous and toxic material (HTM) / B3 category 1. This fact is supported by the XRF analysis of lead-slag founded at site containing high level of Pb, Fe, and Mn (10.027 ppm, 36.94%, and 0.37%) hence the same category is attributed to lead-slag. Conversely, soil in residential areas besides meeting WHO standards (400 ppm), is also categorized as harmless and can be used as a base layering soil for remediation.

Table 4. TCLP Test and Pb-Fractions of Soils and Pb-slag

Sample	Total Pb (ppm)	F1(%)	F2(%)	F3(%)	F4(%)	TCLP Pb(ppm)	Criteria (IGR 101-2014)
Ex-Smelter soil	11.192	25.1	65.2	4.4	5.3	56,2	HTM 1st category
Pb-slag	10.027	36.8	56.5	2.9	3.8	70,9	HTM 1st category
Residential soil	64	45.4	13.6	18.2	22.7	0,17	Non HTM

Table 5. Soil Indices of Contamination Degree and Ecological Risks

Geoaccumulation Index, I_{geo}			Contamination factor, CF		
Value	Category	Description	Value	Category	Description
8.54	$I_{geo} > 5$	Extremely polluted	699.6	CF > 6	Very high contamination
Potential Ecological Risk Factor, E_r			Risk Assessment Code, RAC		
Value	Category	Description	Value	Category	Description
1,749	ER e” 320	Very high risk	90.3	%RAC > 50	Very high risk

Table 4 showed that soil samples are mostly composed of acid-soluble and reducible fraction of Pb(F1 and F2). These two fractions are considered as active/mobile soil-lead fractions. From Table 5, it can be inferred that soil from ex-smelter is extremely – very highly contaminated with lead. The soil also has the highest category of both contamination level and ecological risk. Supporting criteria from Government Regulation 101-2014 (HTM-1), risk assessments based on total concentration (E_r) and geochemical fraction (RAC) also suggested the level of very high risk (VHR) hence the soil needed further treatments.

Designing suitable biochar to remediate the contaminated soil

The contaminated soil possesses the high lead level (11,192 ppm, mostly in mobile fraction F1, F2) and acidic in nature (pH 5.12). This concentration of Pb is equivalent to 0.054M, the level which is high enough in soil needed an effective retainment control such as precipitation (Salam, 2017). In search of suitable biochar, its production, properties, and efficacy in Pb remediation, the results of researchers are summarized in Table 6.

Table 6 shows that biochar having good pores with high surface area produced from natural feedstuff such as rice husks, bamboo, and bagasse is not effective in immobilizing of lead especially at high Pb level (Puga *et al.*, 2015; Rizwan *et al.*, 2016). On contrary, biochar which has precipitating anions such as phosphate, carbonate, sulfate usually produced from mineral-rich materials such as manure and bone is more effective (Liang *et al.*, 2014; Poucke *et al.*, 2019; Himawan *et al.*, 2020). Phosphate is the most effective precipitating agent for lead that forms stable precipitate Hydroxyapatite ($K_{sp} \sim 10^{-67}$) so that the lead concentration remaining in soil solution is very low. Acid condition in soil tends to solubilize lead and increasing one pH unit can reduce soluble Pb up to 118% (Andra *et al.*, 2011). An effective biochar in precipitating Pb having moderate alkalinity will be suitable in the remediation. Factors to be taken into account in producing such biochar are raw material, pyrolysis temperature, particle size, and biochar activation.

High mineral (P, Ca, K) content materials such as manure (poultry, swine, dairy, chicken, cow) and bones should be chosen to produce calcium and phosphate-rich biochar because phosphates are the most stable lead compound in soil (Poucke *et al.*, 2019; Salam, 2017). In the range of 300 – 700 °C, the higher pyrolysis temperature increases ash content and alkalinity of biochar but reduces available phosphates. The choice of pyrolysis temperature is crucial because alkalinity and available phosphate exhibit contradictory effect on Pb mobilityin soil. A moderate temperature (400 - 450 °C) is ideal for pyrolysis. Grinding biochar into finer particle size will increase its contact surface area and sorption capacity. Although increasing pore volume or surface area, washing or activation of biochar is not crucial in immobilization of Pb, Ni, and Cd (Uchimiya *et al.*, 2010; Park *et al.*, 2013). Activation is not recommended in this case because the targeted

Table 6. Bichar production, properties, and performance in Pb immobilization

Author	Material/ Temp./ time	Soil-Pb (ppm), Metal(loid)s	Biochar: pH/Area/ PO_4/ Others	%Dose; Pbreduction	Note
Puga *et al.*, 2015	Sugar cane straw/ 700	141; Cd, Zn; pH 6.1	10.0; 5.0; 900; Ca7,700/ K11,700/S1,900/ash 13.4%	1.5-5; 50%	Low minerals
Liang *et al.*, 2014	Dairy manure/ 350/ 4h	60.7; Cd, Zn, pH7.2	9.1/ -/ 6,400	5; 97.4% minerals	High
Rizwan *et al.*, 2016	Rice straw/	400; Cu, pH5.2	11.2/39.9	3-6; 70%	Low minerals
Domingues *et al.*, 2017	Chicken manure/ 350 450 700	-	9.7-11.7/ - / - / ash 52-54%/ S	0.29-0.44	- High minerals
Poucke *et al.*, 2019	Cow manure/ 550/ 10 min.	17,704; pH4.99	10.76/ - /22,400/	4; 99%	High minerals
Himawan *et al.*, 2020	Chicken (bone, manure) 450	4,029, pH 5.5	8.3; 8.0/ / 10.54; 7.08%/ Ca 34; 18%	5; 79,71; 64.03	High minerals

remediated-soil has minor organic pollutant which needs porous solid absorber.

CONCLUSION

The results showed that the soil in the former recycling center contained Pb above 11,000 ppm which was dominated by reducible fraction (F2) and acid soluble fraction (F1) having extreme contamination level and very high environmental risk. Vertically until 45 cm depth, the deeper the soil the lower the Pb content. Soil and ground water in residential area of Cinangka did not show any lead contamination. Biochar derived from mineral-rich material pyrolyzed at lower temperature is recommended to remediate the contaminated site.

ACKNOWLEDGEMENT

Thanks to Lembaga Pengelola Dana Pendidikan (LPDP) Kementerian Keuangan Republik Indonesia for funding of this research work Contract Number PRJ-4940/LPDP.3/2016.

REFERENCES

Adryansyah, A., Hartono, J.M. and Said, C.A.A. 2019. Remediation of Land Contaminated from Illegal Used Battery Smelting Activity in Cinangka Village, Bogor. *Indonesian Journal of Environmental Education and Management.* 4: 1 DOI: *doi.org/10.21009/jgg.041.01* (in Indonesia)

Ahmad, M., Lee, S. S., Lee, S. E., Al-Wabel, M. I., Tsang, D. C. W. and Ok, Y. S. 2016. Biochar-induced changes in soil properties affected immobilization/mobilization of metals/metalloids in contaminated soils. *Journal of Soil and Sediments.* DOI 10.1007/s11368-015-1339-4

Aktaruzzaman, M., Chowdhury, M.A.Z., Fardous, Z., Alam, M.K., Hossain, M.S. and Fakhruddin, A.N.M. 2014. Ecological Risk Posed by Heavy Metals Contamination of Ship Breaking Yards in Bangladesh. *International Journal of Environmental Research.* 8 (2) : 469-478.

Andra, S.S., Saskar, D., Saminathan, S.K. and Datta, R. 2011. Predicting potentially plant-available lead in contaminated residential sites. *Environ Monit Assess.* 175 : 661-676

Blacksmith Institute, 2014. Project Completion Report Lead Encapsulation Cinangka, Indonesia May 2014.

Cao, X., Ma, L., Liang, Y., Gao, B. and Harris, W. 2011. Simultaneous Immobilization of Lead and Atrazine in Contaminated Soils Using Dairy-Manure Biochar. *Environmental Science & Technology*. Dx DOI.org/10.1021/es103752 u

Himawan, Setyono, P., Suntoro, S. and Rahardjo, S.B. 2020. Characterization and performance of low cost amendments to immobilize lead in contaminated soil. *IOP Conf. Series: Material Sciences and Engineering*. 858012012. DOI: 10.1088/1757-899x/858/1/012012

Huang, G., Su, X., Rizwan, M. S., Zhu, Y. and Hu, H. 2016. Chemical immobilization of Pb, Cu, and Cd by phosphate materials and calcium carbonate in contaminated soils.*Environ Sci Pollut Res.* 23: 16845-16856. DOI 10.1007/s11356-016-6885-9

Imeri, R., Kullaj, E. and Millaku, L. 2019. Distributions of Heavy Metals in Apple Tissues Grown in the Soils of Industrial Area. *Journal of Ecological Engineering.* 20 (3) : 57-66.

Khalid, S., Shahid, M., Niazi, N. K., Murtaza, B., Bibi, I. and Dumat, C. 2016. A comparison of technologies for remediation of heavy metal contaminated soils. *Journal of Geochemical Exploration*, Elsevier. http://dx.doi.org/10.1016/j.gexplo.2016.11.021

Liang, Y., Cao, X., Zhao, L. and Arellano, E. 2014. Biochar- and Phosphate-induced Immobilization of Heavy Metals in Contaminated Soil and Water: Implication of Simultaneous Remediation of Contaminated Soil and Groundwater. *Environ Sci Pollut Res.* 21 : 4665-4674.

Lim, J. E., Ahmad, M., Lee, S. S., Shope, C. L., Hashimoto, Y., Kim, K. R., Usman, A. R. A., Yang, J. E. and Ok, Y. S. 2013. Effects of lime-based waste materials on immobilization and phytoavailability of cadmium and lead in contaminated soil. *Clean - Soil, Air, Water.* 41(12): 1235-1241.

Lu, K., Yang, X., Gielen, G., Bolan, N., Ok, Y. S., Niazi, N. K., Xu, S., Yuan, G., Chen, X., Zhang, X., Liu, D., Song, Z., Liu, X. and Wang, H. 2017. Effect of bamboo and rice straw biochars on the mobility and redistribution of heavy metals (Cd, Cu, Pb, and Zn) in contaminated soil. *Journal of Environmental Management.* 186 : 285-292. http://dx.doi.org/10.1016 /j.jenvman.2016.05.068

Mahar, A., Wang, P., Li, R.H. and Zhang, Z.Q. 2015. Immobilization of lead and cadmium in contaminated soil using amendments: A review. *Pedosphere.* 25 (4): 555-568.

Nemati, K., Abu Bakar, N.K., Abas, M.R. and Sobhanzadeh, E. 2011. Speciation of heavy metals by modified BCR sequential extraction procedure in different depths of sediments from Sungai Buloh, Selangor, Malaysia. *Journal of Hazardous Materials*. 192 : 402-410. http://doi: 10.1016/j.jhazmat.2011.05.039

O'Connor, D., Peng, T., Zhang, J., Tsang, D. C. W., Alessi, D. S., Shen, Z., Bolan, N. S. and Hou, D. 2018. Biochar application for the remediation of heavy metal polluted land: A review of in situ field trials. *Science of the Total Environment*. 619-620: 815-826.

Orillena, E., Custodio, M., Bastos, M.C. and Cuadrado, W. 2019. Lead in Agriculuturalo Soils and Cultivated Pastures Irrigated with River Water Contaminated by Mining Activity. *Journal of Ecological Engineering.* 20 (8): 238-244

Park, J. H., Choppala, G., Lee, S. J., Bolan, N., Chung, J. W. and Edraki, M. 2013. Comparative Sorption of Pb and Cd by Biochars and Its Implication for Metal Immobilization in Soils.*Water Air Soil Pollut.* 224 : 1171. http://doi: 10.1007/s11270-013-1711-1.

Poucke, R.V., Egene, C.E., Allaert, S., Lebrun, M., Bourgerie, S., Morabito, D., Ok, Y.S., Ronsse, F., Meers, E. and Tack, F.M.G. 2019. Application of biochars and solid fraction of digestatento decrease soil solution Cd, Pb and Zn concentrations in contaminated sandy soils. *Environ Geochem Health.* https://doi.org/10.1007/s10653-019-00475-4

Puga, A. P., Abreu, C. A., Melo, L. C. A. and Beesley, L. 2015. Biochar application to a contaminated soil reduces the availability and plant uptake of zinc, lead, and cadmium. *Journal of Environmental Management.* 159 : 86-93.

Rizwan, MS., Imtiaz, M., Huang, G., Chhajro, MA., Liu, Y., Fu, Q., Zhu, J., Ashraf, M., Zafar, M., Bashir, S. and Hu, H. 2016. Immobilization of Pb and Cu in polluted soil by superphosphate, multi-walled carbon nanotube, rice straw and its derived biochar. *Environ Sci Pollut Res.* DOI. 10.1007/s11356-016-6695-0

Rodriguez, I., Gomez, R., Sanchez, V. and Azcarate, J.A. 2015. Chemical and plant tests to assess the viability of amendments to reduces metal availability in mine soils and tailings. *Environ Sci Pollut Res.* 23: 6046-6054.

Salam, Abdul K. 2017. *Management of Heavy Metals in Tropical Soil Environment,* Global Madani Press, Bandar Lampung

Sanchez, M.G., Romera, I.G., Szakova, J., Kaplan, L. and Tlustos, P. 2015. The effectiveness of various treatments in changing the nutrient status and bioavailability of risk elements in multi-element contaminated soil. *Environ Sci Pollut Res.* 22 : 14325-14336. DOI 10.1007/s11356-015-4678-1

Sanchez, M.G., Klouza, M., Holeckova, Z., Tlustos, P. and Szakova, J. 2016. Organic and Inorganic Amendment Application on Mercury-polluted Soils: Effects on Soil Chemical and Biochemical Properties. *Environ Sci Pollut Res.* 23 : 14254-14268. DOI 10.1007/s11356-016-6591-7

Shaheen, S. M. and Rinklebe, J. 2015. Impact of emerging and low cost alternative amendments on the (im) mobilization and phytoavailability of Cd and Pb in contaminated floodplain soil. *Ecological Engineering.* 74 : 319-326. http://dx.doi.org/10.1016/j.ecoleng.2014.10.024.

Tytla, M. 2019. Assessment of Heavy Metal Pollution and Potential Ecological Risk in Sewage Sludge from Municipal Wastewater Treatment Plant Located in the Most Industrialized Region in Poland-Case Study.*International Journal of Environmental Research and Public Health*. 16 : 2430.Doi: 10.3390/ijerph 16132430

Uchimiya, M., Lima, I. M., Klasson, K, T., Chang, S., Wartelle, L. H. and Rodgers, J. E. 2010. Immobilization of Heavy Metal Ions (Cu^{II}, Cd^{II}, Ni^{II}, and Pb^{II}) by Broiler Litter-Derived Biochars in Water and Soil. *J. Agric. Food Chem.* 58 : 5538-5544. DOI: 10.1021/jf9044217.

Wiater, J. 2019. Content of Heavy Metals and Their Fractions in Organic Soils of Podlasie. *Journal of Ecological Engineering.* 20 (3) : 179-184

Wuana, R. A. and Okieimen, F.E. 2011. Heavy Metals in Contaminated Soils: A Review of Sources, Chemistry, Risks and Best Available Strategies for Remediation. *International Scholarly Research Network ISRN Ecology*. 1-20.

Yang, K., Zhang, T., Shao, Y., Tian, C., Cattle, S. R., Zhu, Y. and Song, J. 2018. Fractionation, bioacce-ssibility, and risk assessment of heavy metals in the soil of an urban recreational area amended with composted sewage sludge. *International Journal of Environmental Research and Public Health.* 15 : 613. Doi: 10.3390/ijerph 15040613.

Yang, Q., Hu, G., Yu, R., He, H. and Lin, C. 2016. Distribution, fractionation, and contamination assessment of heavy metals in offshore surface sediments from western Xiamen Bay, China. *Acta Geochim.* 35 (4): 355-367

Yang, X., Liu, J., McGrouther, K., Huang, H., Lu, K., Guo, X., He, L., Lin, X., Che, L., Ye, Z. and Wang, H. 2016. Effect of biochar on the extractability of heavy metals (Cd, Cu, Pb, and Zn) and enzyme activity in soil. *Environ Sci Pollut Res.* 23 : 974-984. DOI 10.1007/s11356-015-4233-0

Zhang, L., Liao, Q., Shao, S., Zhang, N., Shen, Q. and Liu, C. 2015. Heavy Metal Pollution, Fractionation, and Potential Ecological Risks in Sediments from Lake Chaohu (Eastern China) and the Surrounding Rivers. *International Journal of Environmental Research and Public Health.* 12 : 14115-14131. Doi: 10.3390/ijerph 121114115.

Zhang, X., Wang, H., He, L., Lu, K., Sarmah, A., Li, J., Bolan, N. S., Pei, J. and Huang, H. 2013. Using biochar for remediation of soils contaminated with heavy metals and organic pollutants. *Environ Sci Pollut Res.* 20 : 8472-8483. DOI 10.1007/s11356-013-1659-0.