

Phytoremediation of Lead (Pb) and Arsenic (As) Contaminated Soil in Artisanal Gold Mining at Selogiri, Wonogiri District, Central Java, Indonesia

Bambang Suryo Madyo Pranoto and Wawan Budianta*

Department of Geological Engineering, Faculty of Engineering, Universitas Gadjah Mada, Yogyakarta, Indonesia

ABSTRACT. Artisanal gold mining (ASGM) is commonly found in Indonesia, particularly in Wonogiri District, Central Java. One of the impacts of ASGM activity is soil contamination influenced by mining waste. This study's objective was to investigate the potential use of *Amaranthus spinosus* L. and *Jatropha curcas* for remediation of lead (Pb) and arsenic (As) in contaminated soil. The study investigated translocation and accumulation of lead (Pb) and arsenic (As) from the soil into the plant's roots and shoots. The phytoremediation experiment was observed using *Amaranthus spinosus* L. and *Jatropha curcas* plants and evaluating both plant's effectiveness as a hyperaccumulator. The result shows that higher Pb and As concentration were found in roots rather than in both plants' shoots. However, *Jatropha curcas* use seems more effective in reducing Pb and As concentrations than *Amaranthus spinosus* L in both shoots and roots. Generally, the use of both hyperaccumulator plants was more effective in Pb remediation compared to As. This phytoremediation experiment revealed *Amaranthus spinosus* L. and *Jatropha curcas* reduces Pb concentrations and As in contaminated soil. This reduction is the critical point leading to the entry of Pb and As into the food chain.

Keywords: Phytoremediation · Artisanal gold mining · *Amaranthus spinosus* L · *Jatropha curcas*.

1 INTRODUCTION

Artisanal small-scale mining (ASGM) activity is commonly found in developing countries like Indonesia (Aspinall, 2001). One of the environmental issues related to ASGM is the heavy metal contamination in the soil, which caused degradation of soil quality (Alloway, 1995). Soil provides plants for their roots and holds the necessary nutrients for plants to grow. It filters the rainwater and regulates the discharge of excess rainwater; it also buffers against pollutants, thus protecting groundwater quality (Doran *et al.*, 2000). Several studies revealed that artisanal mining impacts metals contamination in soil around the gold mining processing area (Ap-

leton *et al.*, 2006; Molina *et al.*, 2006). The metals contaminated soil samples were obtained from the ASGM area in Wonogiri District, Central Java, Indonesia (Figure 1).

Geologically, the ASGM was located on the bedrock consist of tuff, andesite, diorite-microdiorite intrusion. The ore deposits types in this area are typical of low sulphidation epithermal mineralization system with the presence of pyrite (FeS₂), chalcopyrite (CuFeS₂), galena (PbS), and arsenopyrite (FeAsS) (Soe *et al.*, 2005). In this area, the level of arsenic contamination is well documented in a previous study (Harijoko *et al.*, 2010). One of the cost-effective soil remediation methods is phytoremediation as a plant-based approach of remediation that takes advantage of plants' ability to accumulate heavy metals elements and compounds from the soil and metabolize

*Corresponding author: W. BUDIANTA, Department of Geological Engineering, Universitas Gadjah Mada. Jl. Grafika 2 Yogyakarta, Indonesia. E-mail: wbudianta@ugm.ac.id

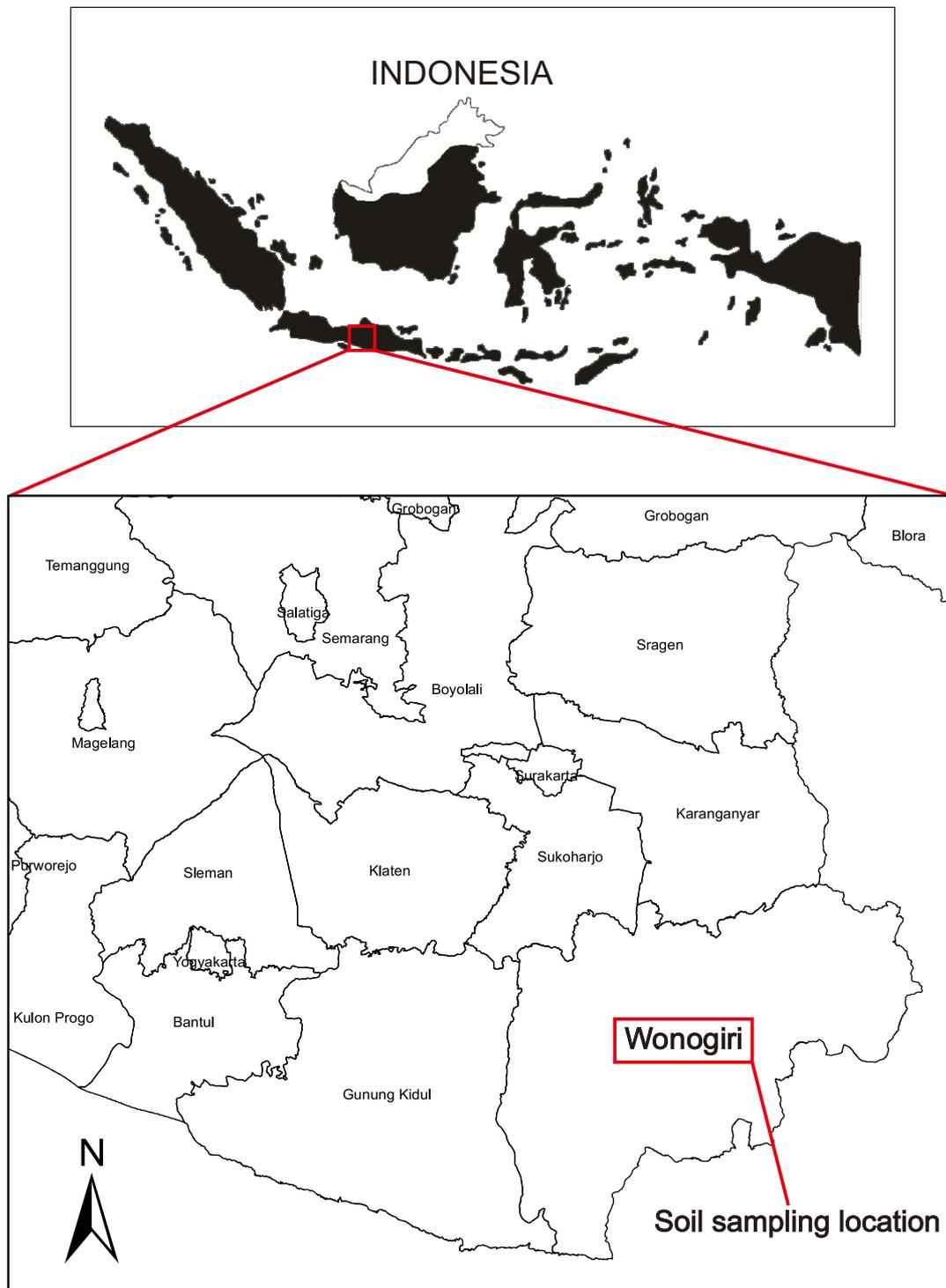


FIGURE 1. Location of artisanal gold mining and contaminated soil sampling point.

various molecules' plant's tissues (Raskin and Ensley, 2000). Several works have been conducted to investigate the potential use of *Amaranthus spinosus* L. and *Jatropha curcas* for the hyperaccumulator plant of phytoremediation studies (Chang *et al.*, 2014; Huang *et al.*, 2019). This study aims to investigate the *Amaranthus spinosus* L. and *Jatropha curcas* for remediation of Pb and As in contaminated soil obtained from the ASGM area.

2 MATERIAL AND METHODS

Mining waste (tailing) as the primary Pb source and As was sampled and then measured its concentration. The vein or ore mineral in the gold mining area was sampled and measured for Pb and As concentration to understand the natural background. Contaminated soil samples for this study were obtained from the most polluted sites near the gold processing area using a hand auger made of stainless steel (Figure 2). The soil samples were obtained from three layers: top layer (5 cm), middle layer (10 cm), and low layer (15 cm). The samples from a given layer were collected to ensure that the result would represent contaminated soil in the study area. The soil samples were placed in plastic boxes. They were transported to the laboratory, where they were dried at room temperature to constant weight. The soil samples then refined in an agate mortar. The soil samples were then collected and sieved through a sieve of mesh size 0.150 mm. The total concentrations of the Pb and As were measured by Inductively coupled plasma atomic emission spectroscopy (ICP-AES). The phytoremediation experiment was conducted by assessing *Amaranthus spinosus* L. and *Jatropha curcas* in Pb and As contaminated soil samples.

As mentioned in the previous section, the Pb and As contaminated soil were obtained from the gold processing area (Figure 2). For the experiments, the plastic bag was used containing 2 kg of soil, placed in a greenhouse. The phytoremediation experiment was carried out under greenhouse conditions to understand better the patterns of response, distribution, and accumulation of heavy metals in plants. The advantage of a greenhouse is securing a reasonable amount of maintained heat, water vapors, and humidity (Datta *et al.*, 2011). Contaminated soil

samples were obtained from the ASGM area in three depth (soil A with 5 cm depth) (soil B with 10 cm depth) and (soil C with 15 cm depth), as shown in Figure 3. Control plastic bags with uncontaminated soil were also prepared (Figure 3). From each pot, three different subsets of by *Amaranthus spinosus* L. and *Jatropha curcas* were harvested at three different times each month during the growing season for three months.

The plants were split into shoots (as a combination of stem and leaves) and roots, thoroughly washed with distilled water to remove adhering soil particles and dried at 35°C to constant weight. Plant material was then milled, weighed, and determined of Pb and As concentration by ICP AES after digested in 25 ml aqua regia.

3 RESULTS AND DISCUSSION

3.1 Pb and As in soil samples

The result of Pb and As concentrations on the samples of vein or minerals and tailings samples were shown in Table 1. As shown in Table 1, the Pb and As in tailing level was higher than the mean crust average stated by the previous study (Wedepohl, 1995). The laboratory test results showed Pb and As concentration in 3 soil samples had values 356, 212, 98 ppm and 10.8, 10.5, 7.8 ppm of soil with depth 5 cm, 10 cm, and 15 cm respectively for Pb and As. In summary, the Pb and As the concentration of the data is presented in Table 1.

Table 1 shows that the concentration of Pb and As in the soil sample obtained from the ASGM area was higher than the mean crust average or natural condition (Wedepohl, 1995). The control soil used in this experiment shows that Pb concentration and As in the range of natural concentration. Generally, all soil samples have a low content of silt-clay fraction, and the type was loamy fine sand soil according to the United States Department of Agriculture (USDA) classification. Organic content in the soil samples relatively low and reflects on their CEC value.

3.2 Phytoremediation experiment

Figure 4 shows the phytoremediation experiment during the three-month observation. It



FIGURE 2. The field condition of soil sampling.

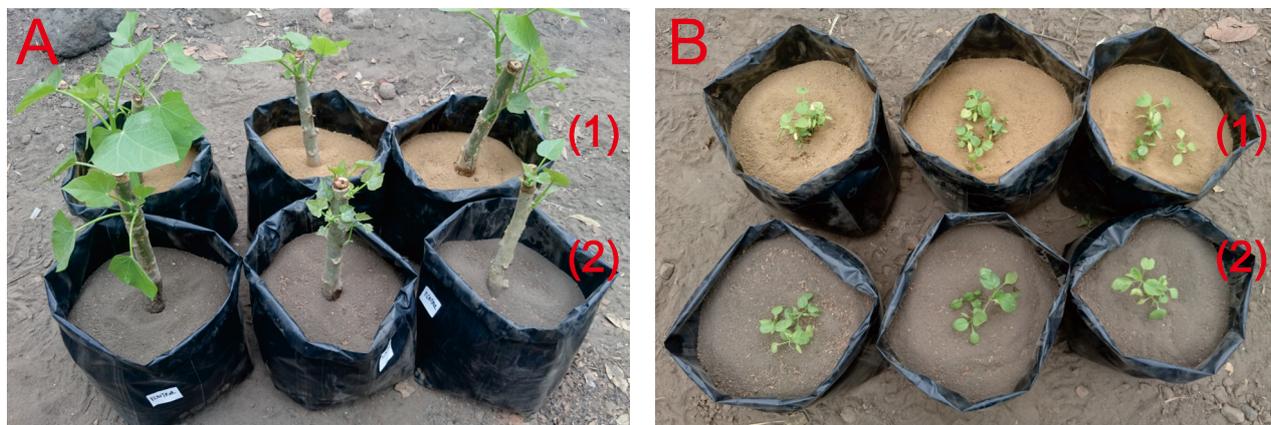


FIGURE 3. Initial plant on the experiment for *Jatropha curcas* (A), *Amaranthus spinosus* L. (B), and (1) contaminated soil and (2) control soil.

TABLE 1. Results of Pb and As concentration measurement of samples.

No.	Sample	Pb (ppm)	As (ppm)	CEC (meq/100 gr)	Grain Size (% silt-clay)	Organic Content (%)
1	Veins/mineral	562	190	-	-	-
2	Tailing	480	170	-	-	-
3	Soil (5 cm depth)	356	10.8	14	15	5
4	Soil (10 cm depth)	212	10.5	12	18	3
5	Soil (15 cm depth)	98	7.8	14	16	3
6	Control soil	18	1.8	9	14	2
7	Mean crust average*	16	1.8	-	-	-

* Wedepohl, 1995

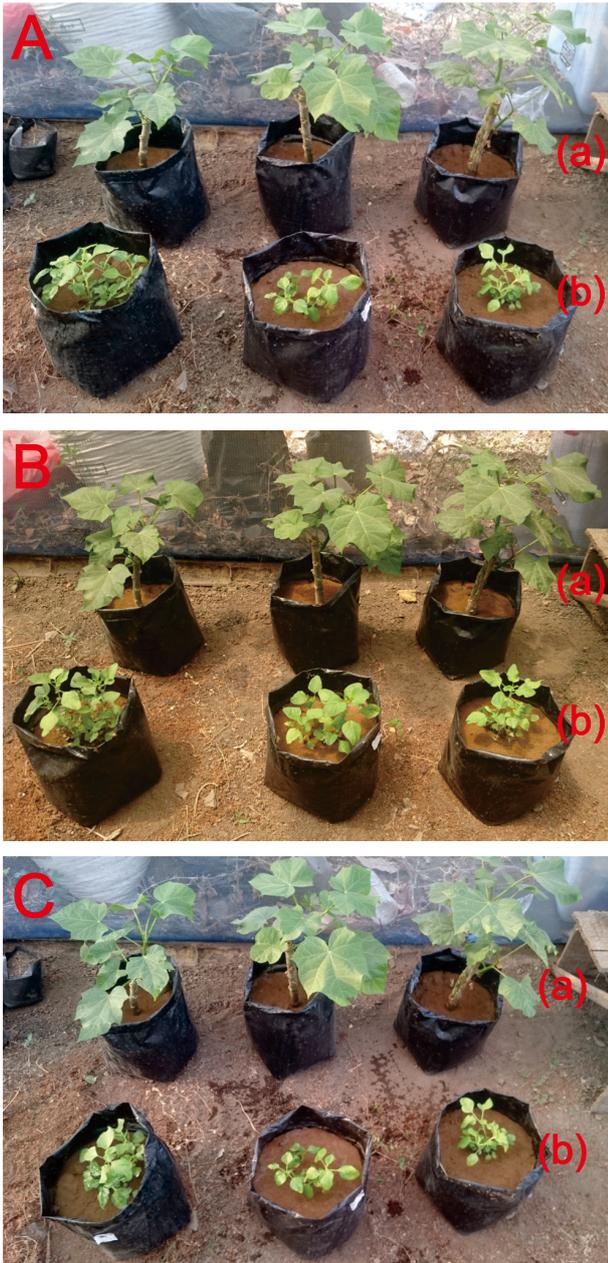


FIGURE 4. *Jatropha curcas* (a) and *Amaranthus spinosus* L. (b) with harvesting time 1 (A), harvesting time 2 (B), and harvesting time 3 (C).

shows the growth of stem and leaves of *Jatropha curcas* and *Amaranthus spinosus* L. in three harvesting times.

Tables 2 and 3 summarize the Pb and As concentrations in the shoots and roots in the experiment. Generally, the relative Pb and As absorption by plant tissues is higher in roots than shoots. The results of Pb and As concentration (Tables 2 and 3) show a percentage concentration of more than 400% in shoots and 1000% in roots relative to the controls when the

plant was applied on contaminated soil A. The lowest concentrations of Pb and As in *Jatropha curcas* shoots were obtained in first harvesting. There are significant increases in Pb concentrations, and As in *Jatropha curcas* roots, the increase was evident at second and third harvesting. The percentage Pb and As concentration compared to the controls are less than for the shoots.

The data in Figure 5–8 show a progressive increase in Pb and As concentrations with time. For example, the Pb concentration in *Jatropha curcas*' roots has 81 ppm in the first harvesting time and increases to 92 ppm during the end of harvesting in soil A (Figure 4). A similar pattern also obviously for *Amaranthus spinosus* L. for soil A. The concentration on the roots has 75 ppm in the first harvesting time and increases to 79 ppm during harvesting.

Tables 2 and 2 show that Pb and As concentrations in *Jatropha curcas* were higher than *Amaranthus spinosus* L. The higher concentration is found in both shoots and roots, similar to finding with another study (Chang *et al.*, 2014; Álvarez-Mateos *et al.*, 2019). The higher concentration of metals is definite in the shoots than in the *Jatropha curcas* and *Amaranthus spinosus* L. In this phytoremediation study, Pb and As concentrations were higher in the roots than in the shoots is confirmed by other studies. Generally, a trend for Pb and As to concentrate in roots demonstrates that the roots are delivered as a barrier to Pb and As uptake (Majid *et al.*, 2012; Ziarati and Alaedini, 2014). This phenomenon occurs because the roots' heavy metal uptake is more rapidly compared with the transport to other plant tissues. Generally, the use of both hyperaccumulator plants was more effective in Pb remediation compared to As, in line with the resulting form of another study (Gonzalez-Chavez *et al.*, 2017).

The highest Pb and As concentration in the shoots were found during the third month for both plants. This evidence is due to an increase in the biomass as a function of exposure time, which causes dilution of the Pb and As concentration in a process similar to that which occurs in the shoots. Moreover, the shoots' primary function is to conduct nutrients to other parts of the plant and not accumulate them. Also, Pb and As accumulation in the stems and leaves

TABLE 2. Summary of the experimental result of Pb contaminated soil.

Plant/harvest time	<i>Jatropha curcas</i>				<i>Amaranthus spinosus L.</i>				
	Control ¹	A ²	B ³	C ⁴	R% ⁵	A ²	B ³	C ⁴	R% ⁵
Shoots									
Harvest time (1)	5	26	23	8	520.0	18	16	2	320.0
Harvest time (2)	5.8	27	21	9	465.5	20	17	2	293.1
Harvest time (3)	6.5	29	22	13	446.2	21	20	3	307.7
Roots									
Harvest time (1)	10	81	67	65	810.0	75	56	45	560.0
Harvest time (2)	12.8	84	75	62	656.3	76	63	48	492.2
Harvest time (3)	12.9	92	83	76	713.2	79	66	51	434.1

¹ control soil

² plant in contaminated soil in 10 cm depth

³ plant contaminated soil in 15 cm depth

⁴ plant in contaminated soil in 20 cm depth

⁵ percentage concentration of Pb in the plant of soil A relative to control.

TABLE 3. Summary of experimental result of As contaminated soil.

Plant/harvest time	<i>Jatropha curcas</i>				<i>Amaranthus spinosus L.</i>				
	Control ¹	A ²	B ³	C ⁴	R% ⁵	A ²	B ³	C ⁴	R% ⁵
Shoots									
Harvest time (1)	0.3	12	5	5	1500.0	7	1	1	1125.0
Harvest time (2)	0.7	15	6	5	1250.0	8	2	1	666.7
Harvest time (3)	0.8	16	9	7	1230.8	9	5	5	538.5
Roots									
Harvest time (1)	0.7	54	41	14	4909.1	23	8	4	2090.9
Harvest time (2)	0.8	56	43	20	4666.7	20	8	5	1666.7
Harvest time (3)	0.9	60	51	24	5000.0	14	9	8	1166.7

¹ control soil

² plant in contaminated soil in 10 cm depth

³ plant contaminated soil in 15 cm depth

⁴ plant in contaminated soil in 20 cm depth

⁵ percentage concentration of As in the plant of soil A relative to control.

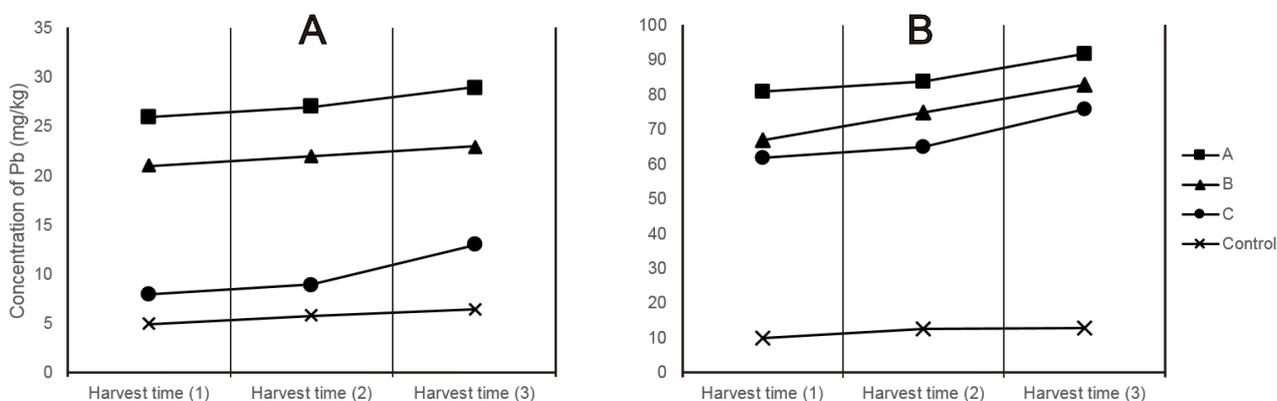


FIGURE 5. Concentration Pb *Jatropha curcas* in the shoot (A) and root (B).

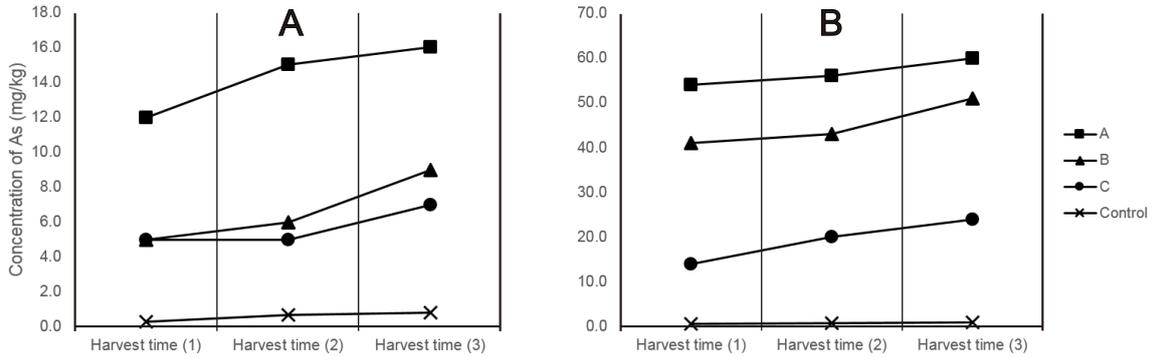


FIGURE 6. Concentration As *Jatropha curcas* in the shoot (A) and root (B).

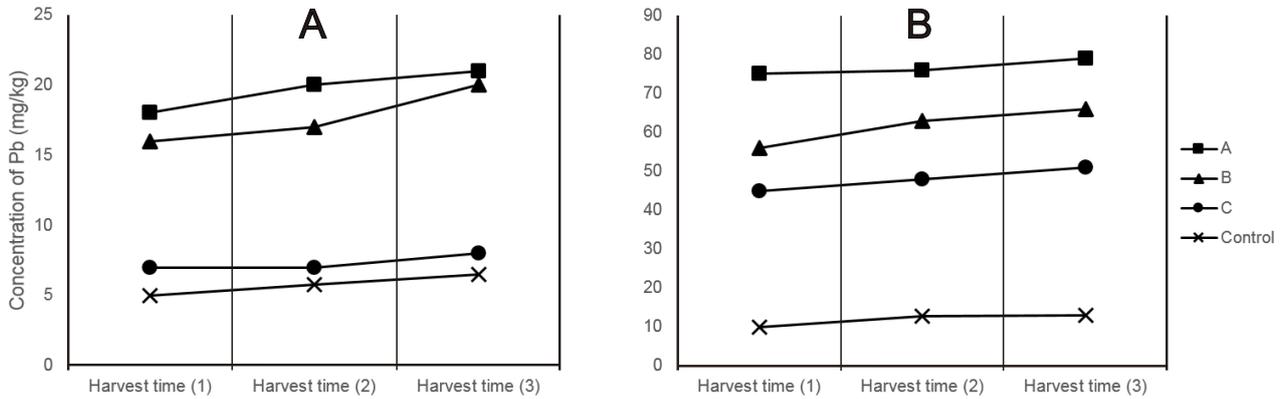


FIGURE 7. Concentration Pb *Amaranthus spinosus L.* in the shoot (A) and root (B).

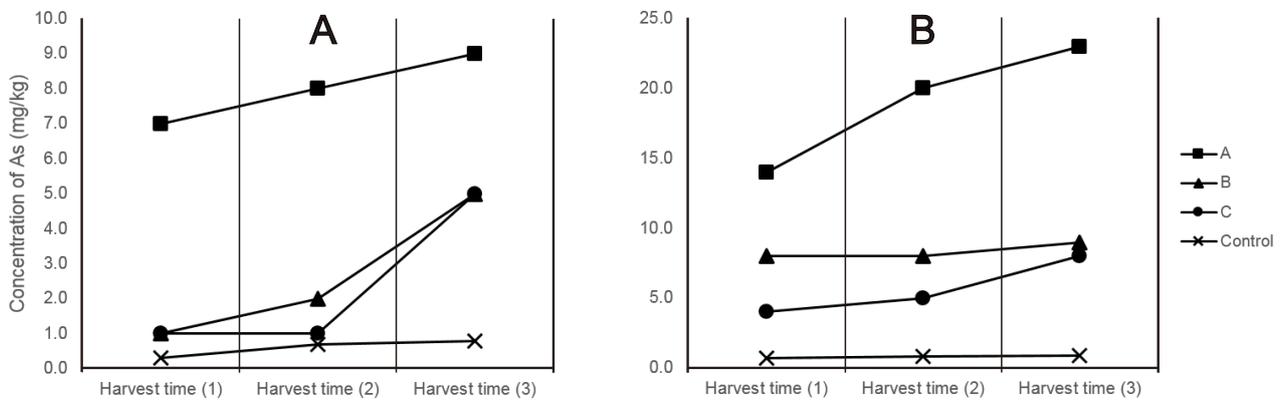


FIGURE 8. Concentration As *Amaranthus spinosus L.* in the shoot (A) and root (B).

(shoots) are limited compared with the other plant tissues due to the high retention of Pb and As by the roots. These factors result in a lower Pb and As concentration in shoots than in roots. The results showed that the maximum accumulating value on roots was about 92 and 60 ppm on a dry weight basis for Pb and As respectively (see Tables 2 and 3). Based on these results, *Jatropha curcas* is likely an excellent candidate to be considered a potential Pb and As accumulation plant species.

4 CONCLUSION

Several findings can be obtained from the present study. The phytoremediation experiment shows that the higher Pb and As concentration was found in roots rather than shoots in the *Jatropha curcas* and *Amaranthus spinosus* L. From two plants used in this study, the accumulation Pb and As concentration seems to be higher in *Jatropha curcas* compared to *Amaranthus spinosus* L. Even though the Pb and As levels uptake by the shoots were relatively low, *Jatropha curcas* is a suitable choice for the phytoremediation of Pb and As contaminated soil because it is easy to plant and maintain and contributes to soil remediation and this species commonly found around gold mining areas. The most crucial factor controlled the Pb and As removal in contaminated soil is the exposure time. The results of this study expand the knowledge of the Pb and As absorption and translocation capacity of *Jatropha curcas* and *Amaranthus spinosus* L.

ACKNOWLEDGEMENTS

The authors gratefully thank Gadjah Mada University for its financial support to this research work, Tokyo Institute of Technology for its ICP-AES to this sample analysis, the Village Head of the Jendi for his hospitality during sampling, and M. Eka Danta Winsu Branta for his technical support and use of greenhouse spaces. The authors also thank the Editor and reviewers whose valuable suggestions improved the content of this manuscript.

REFERENCES

Alloway B.J. (1995) Heavy Metals in Soils. 2nd edition. Blackie and Sons, Glasgow.

- Aspinall C. (2001) Small-scale mining in Indonesia. International Institute for Environment and Development, Mining Minerals and Sustainable Development Report, Jakarta.
- Álvarez-Mateos P., Alés-Álvarez F.J., García-Martín J.F. (2019) Phytoremediation of highly contaminated mining soils by *Jatropha curcas* L. and production of catalytic carbons from the generated biomass, *Journal of Environmental Management* 231, 886-895.
- Appleton, J.D., Weeks, J.M., Calvez, J.P.S., Beinhoff, C. (2006) Impacts of mercury contaminated mining waste on soil quality, crops, bivalves, and fish in the Naboc River area, Mindanao, Philippines, *Science Total Environment* 354, 198-211.
- Chang, F.C., Ko, C.H., Tsai, M.J., Wang, Y.N., Chung C.Y. (2014) Phytoremediation of heavy metal contaminated soil by *Jatropha Curcas*, *Ecotoxicology* 23:1969-1978.
- Datta, R., Quispe, M. A., Sarkar, D. (2011) Greenhouse study on the phytoremediation potential of vetiver grass, *Chrysopogon zizanioides* L., in arsenic-contaminated soils, *Bulletin of environmental contamination and toxicology*, 86(1), 124-128.
- Doran, J.W. and Zeiss, M.R. (2000) Soil health and sustainability: managing the biotic component of soil quality, *Applied Soil Ecology* 15, 3-11.
- Gonzalez-Chavez, M.D.C.A., Carrillo-Gonzalez, R., Hernandez Godinez, M.I., and Evangelista Lozano, S. (2017) *Jatropha curcas* and assisted phytoremediation of a mine tailing with bio-char and a mycorrhizal fungus, *International Journal of Phytoremediation*, 19, 174-182.
- Harijoko, A., Htun, T.M., Saputra, R., Warmada, I W., Setijadji, L.D., Imai, A., Watanabe, K. (2010) Mercury and arsenic contamination from small scale gold mining activities at Selogiri area, Central Java, Indonesia. *Journal of Southeast Asian Applied Geology* 2: 56-69.
- Huang Y., Xi Y., Gan L., Johnson D., Wu Y., Ren D., Liu H. (2019) Effects of lead and cadmium on photosynthesis in *Amaranthus spinosus* and assessment of phytoremediation potential. *International journal of phytoremediation*, 21(10), 1041-1049.
- Majid, N.M., Islam, M.M., and Riasmi, Y. (2012) Heavy metal uptake and translocation by *Jatropha curcas* L. in sawdust sludge contaminated soils, *Australian Journal Crop Science* 6(5), 891-898.
- Molina, J.A., Oyarzun, R., Esbrí, J.M., Higuera, P. (2006) Mercury accumulation in soil and plants in the Almadén mining district, Spain, one of the most contaminated sites on Earth, *Environmental Geochemistry and Health* 28, 487-498.
- Raskin, I. and Ensley, B.D. (Eds) (2000) *Phytoreme-*

- diation of toxic metals: using plants to clean up the environment. John Wiley and Sons, Inc.
- Soe, M. T. (2005) Geology and Gold-Copper mineralization at Selogiri area, Wonogiri Regency, Central Java, Indonesia, Gadjah Mada University, Yogyakarta, Indonesia, Master Degree Thesis (unpublished).
- Wedepohl, K.H. (1995) The composition of the continental crust, *Geochimica et Cosmochimica Acta* 59 (7), 1217–1232.
- Ziarati P., Alaedini S. (2014) The Phytoremediation Technique for Cleaning up Contaminated Soil by *Amaranthus sp*, *Journal of Environmental Analytical Toxicology* 4:208.