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The Potential of Local Trees for Phytostabilization of Heavy Metals in Gold Cyanidation Tailing Contaminated Soils of West Lombok, Indonesia

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ABSTRACT

The discharge of mercury amalgamation and cyanidation tailings of small-scale gold mining in West Lombok, to agricultural lands caused Hg, Cd, Cu, Pb and Zn contamination. Phytostabilization may be a feasible approach for the management of contaminated sites. The objective of this study was to search for tree species that are potential for phytostabilization of soils contaminated by small-scale gold mine tailings in West Lombok, Indonesia. Results of this study showed that **t**here were at least 28 tree species found in areas contaminated by gold cyanidatation tailing. Based on cyanide concentration, Hg concentration, and energy of above-ground parts of the identified plants, *Duabanga moluccana* (DM), *Erythrina orientalis* (DM)., and *Paraserianthes falcataria* (PF) were selected to study their phytostabilization potential. Results of plant growth experiment showed that the tolerance of the three species to heavy metals was in the order of Zn > Pb > Cu > Cd. The highest concentration of Cd (0.9 mg/kg) was found in the roots of DM. The highest concentration of Cu (37,7 mg/kg) was found in roots of EO. Roots of PF contained the highest concentrations of Pb (37.7 mg/kg) and Zn (546 mg/kg). The three plant species had metal shoot / metal root ratios of more than one. This indicates that *Duabanga moluccana, Paraserianthes falcataria*, and *Erythrina orientalis* are suitable for phytostabilization of Cu, Pb, Cd, and Zn in gold cyanidation tailing contaminated soils of West Lombok, Indonesia.

Keywords: cadmium, copper, lead, phtoremediation, trees, zinc

INTRODUCTION

The gold mining sector in Indonesia consists of large-scale, medium-scale, and artisanal and small-scale gold mining (ASGM). Indonesia is regarded as a major location for ASGM activities. However, few monitoring or new-technology demonstration projects appear to have been conducted in Indonesia. Aspinall [1] reported that there are 713 illegal small-scale mining sites throughout Sumatra, Java, Kalimantan and Sulawesi, with the majority of them ASGM. At many ASGM in Indonesia, a two-stage process of amalgamation followed by cyanidation is used to ensure maximum recovery of gold from ore [23]. One of them is located at Sekotong District of West Lombok. In this area, rock is removed by hand from simple mine shafts, and ground the rock in simple ball grinders (local name 'gelundung'). Mercury is added during the later stages of grinding, and the amalgam is panned off. The amalgam is heated using gas burners releasing volatile mercury into the atmosphere. A large proportion of the amalgamation tailings are sold to cyanidation plants that area far away from the amalgamation sites. Tailings are placed in leaching tanks, and an alkaline solution of sodium cyanide circulated to dissolve residual gold. After a prescribed leach time, the gold-rich cyanide solution is filtered through activated carbon. This removes the gold from solution. The carbon is subsequently washed and burned. Gold in the ash is collected using a final amalgamation step. Cyanide tailings are then discharged directly into adjacent land. At one facility in the Sekotong district cyanidation tailings are drained directly from the leach tank into a rice paddy behind the cyanidation processing facility. The volume of tailings in the rice

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paddy is increasing with each leach, as is the spatial extent of the distribution of the cyanidation waste. A field survey conducted earlier by Prasetya *et al*. [16] showed that the chemical characteristics of the deposited cyanidation tailing were as follows: sandy loam texture, pH 7.7, 1.19% organic-C, 0.001% total N, 2.89 mg total P/kg; 1.27 mg S/kg; 11.57 cmol/kg CEC; 792 mg Cu/kg; 4.0 mg Cu/kg; 1,090 mg Hg/kg; 1.68 mg Au/kg; Pb 530 mg/kg; 3,810 mg Fe/kg; 4,840 mg Mn/kg; and 3,760 mg Zn/kg.

Soils contaminated by mine tailings containing heavy metals can be restored using chemical treatments, and/or physical treatments [2,10]. However, such technologies are destructive, expensive, and do not achieve long term solution [4]. A promising sustainable technique for metal remediation is phytoremediation [15]. Phytoextraction and phytostabilization and are the most usual phytoremediation techniques adopted for soils contaminated with heavy metals. Because long-term accumulation of metals in the aboveground biomass of plants may pose a risk of transfer to food chain, phytostabilization may be a more feasible approach for the management of contaminated sites than phytoextraction [12]. Use of native plants is a focus of phytostabilization. As there can be a wide range of indigenous plants that are very adaptive, the best ways to initiate species selection is by observing the indigenous plant species that have grown on disturbed adjacent areas [13]. A plant species can be considered potential for phytostabilzation if accumulation factor (AF) = (total element concentration in shoot tissue: total element concentration in mine tailings) and (b) shoot: root (S: R) ratio = (total element concentration in shoot tissue: total element concentration in the root tissue), are less than one [3]. Trees provide an extensive canopy cover and establish a deeper root network to prevent erosion over the long term [11]. Trees provide a high nutrient environment for grasses while reducing moisture stress and improving soil physical characteristics in arid and semiarid climates [22]. Leguminous trees that serve as a nitrogen supply such as *Acacia* spp. and *Prosopis* spp. have been reported as successfully colonizing mine tailings in the Western United States [5].

The objectives of this study were to search for tree species that are potential for phytostabilization of soils contaminated by small-scale gold mine tailings in West Lombok, Indonesia. This study was initiated by identification of metal-tolerant plant species from natural vegetation in fields contaminated with gold mine tailings. A further study was then conducted to assess the feasibility to use the selected plants for phytostabilization through plant growth study.

Materials and Methods

Site description and Observation of local trees:

Field observation of local trees was carried out in September 2013 at areas adjacent to the tailing disposal area of a cyanidation plant at Sekotong District of West Lombok, Indonesia (115^0.46'-116^0.20'E and 8^0.25'-8^0.55'S). The area is located at 400 m above sea level with average annual rainfall of 1500 mm. In small-scale gold mining of this area, gold-bearing ore or soil was processed using an equipment called 'Gelundung', to separate the gold from the surrounding ore body. The equipment consists of between one to ten cylinders which are powered by diesel engine or waterwheels that mix the gold bearing soil or rock with mercury. Spinning for about 9-12 hours, the cylinders grind the small rocks inside the soil into a raw paste mixture of gold, and mercury, which is then strained through a porous cloth which separates the gold from the mercury. The end product of this process is called bullion. As the tailings can still contain traces of gold, along with used mercury, the tailings were sold at a much lower market price for reprocessing of gold using cyanide with concentration of 600-800 mg/kg. The cyanidation process tailings were then deposited to tailing dams nearby the processing units.

Tree species identification was carried out in the field surrounding the tailing disposal areas using quadrate method of Oosting [8] to calculate basal area (dominancy), density, and frequency of species. Three transects of 50 m x 50 m each with a distance of 40 m were made vertically crossing to the tailing disposal area. Within each transect, 5 plots of 1x1 m dimension were made with interval of 5 m, resulted in 15 plots for the three transects. Every tree species found in each plot was recorded. Dominant tree species was estimated using important value of the species which is the sum of relative density, relative frequency and relative dominancy of the species. Relative density, frequency and dominancy of species are the percentage that the species contributes to the total density, frequency and basal area (dominancy), respectively.

Plant and soil sample analysis:

Above ground parts (twigs and leaves) of tree species having important index values of more 10% were collected, washed with distilled water to adhered soil particles, and oven dried at 60^oC for 48 hours. Energtic species were determined based on their calory that was measured using a bomb calorimeter. Calory of the plant biomass (H Kcal/g) was calculated using the following equation, H = (heat capacity x change in temperature) x $(1000\ m)^3$. Soil samples from the rooting zone (0–15cm) of the fifteen plots were mixed and air-dried at room temperature for two weeks and then sieved by 2-mm sieve. Total cyanide content in plant and soil samples were determined argentometrically using nitrate silver ($AgNO_3$) as a titration standard. A 20 g of dry ground sample was mixed with 10mL H_2SO4 and

100mL distilled water. The distillate was put in a vial containing 20mL of 2.5% NaOH, and then titrated with 0.02N $AgNO_3$ until the solution turned to red. The concentration of Hg in the soil and plant samples was determined using a F732-S Cold Atomic Absorption Mercury Vapor Analyzer (Shanghai Huaguang Instrument Company).

Growth experiment of selected potential plants:

Based on cyanide and Hg concentrations of identified plants, three plants were selected to study their phytostabilization potential. A pot experiment was conducted from October to December 2013 in a glasshouse of the Department of Soil Science, Brawijaya University having average temperature of 27^{o}C and humidity of 93%. Four growing media tested in this study were one tailing uncontaminated soils and three mixtures of tailings and compost (100:0, 80:20, and 60:40% weight). Two pre germinated seeds of each selected plant were planted in each of the twelve planting media. The twelve treatments (three plants and four planting media) were arranged in a completely randomized design with four replicates. The moisture content of the medium in each pot was adjusted to its approximate water holding capacity. Water was supplied daily to each pot in order to keep the moisture content of the tailing medium at the approximate water holding capacity. At every week, until harvest at 8 weeks, plant height was measured. At harvest, plant shoots and roots were separated and then oven-dried at 60^{o}C for 72 hours, weighed and ground to pass through a 1 mm sieve for cyanide and Hg analyses. Data obtained were subjected to analysis variance followed by 5% least significance different test

Results and Discussion

There were at least 28 tree species found in areas contaminated by gold cyanidatation tailing of Sekotong District of West Lombok (Table 1). Amongts them, tewlve tree species were found to have frequancy value of more that 10, i.e. *Aglaia odorata* Lour., *Aquilaria malaccensis* Lam., *Dracontomelon dao* (Blanco) Merr., *Duabanga moluccana* Blum., *Erythrina orientalis* L., *Eugenia subglauca* Koord. & Valeton., *Kleinhovia hospita* L., *Lagerstromeia speciosa* (L.) Pers., *Melochia umbellata* (Houtt.) Stapf., *Paraserianthes falcataria* (L.) Nielsen., *Schleichera oleosa* (Lour.) Oken., and *Toona sureni* (Blume) Merr. (Table 1). Those tree species are secondary pioneer species that survived under nutrient-poor conditions, and thus they were expected to be potential for phytoremediation of heavy metal contaminated soils.

Table 1: Local tree species identified in gold cyanidation tailing contaminated area of Sekotong District of West Lombok.

No	Botanical name	D	C	F	RD	RC	RF	IIV	DI
1.	*Aglaia odorata* Lour.	209	1044	41	6.77	9.01	11.6	27.37	0.323
2.	*Alstonia* scholaris (L.) R.Br.	11	225	4	0.44	2.04	1.09	3.57	0.034
3.	*Anthocephalus* cadamba Miq.	7	100	7	0.28	0.91	1.9	3.09	0.024
4.	*Aquilaria malaccensis* Lam	128	303	17	3.83	2.55	2.01	8.39	0.180
5.	*Artocarpus utilis* L.	3	25	3	0.12	0.23	0.82	1.16	0.012
6.	*Calophyllum soulatri* Burm.f.	11	225	4	0.44	2.04	1.09	3.57	0.034
7.	*Casia siamea* (Lam.) Irwin & Barneby	8	50	5	0.24	0.42	1.44	2.1	0.021
8.	*Dracontomelon dao* (Blanco) Merr.	339	2455	58	11.63	21.28	16.3	49.16	0.477
9.	*Drypetes longifolia* (Blume) Pax	33	23	2	1.32	0.21	0.54	2.07	0.082
10.	*Duabanga moluccana* Blum.	17	160	8	0.68	1.45	2.17	5.31	0.049
11.	*Erythrina orientalis* L.	33	235	10	1.25	2.05	2.78	6.08	0.087
12.	*Eugenia subglauca* Koord. & Valeton	465	606	14	14	5.13	3.99	23.12	0.421
13.	*Garciana celebica* L.	10	70	1	0.4	0.64	0.27	1.31	0.032
14.	*Heritiera littoralis* Dryand.	1	85	1	0.03	0.72	0.29	1.03	0.004
15.	*Intsia bijuga* (Colebr.) Kuntze.	5	10	2	0.2	0.09	0.54	0.83	0.018
16.	*Kleinhovia hospita* L.	241	639	32	8.12	5.56	8.91	22.59	0.375
17.	*Lagerstromeia speciosa* (L.) Pers.	121	413	11	3.63	3.49	3.14	10.27	0.177
18.	*Manilkara kauki* (L.) Dubard	30	84	6	1.15	0.75	1.68	3.57	0.08
19.	*Melochia umbellata* (Houtt.) Stapf.	241	2620	41	8.56	22.96	11.4	42.95	0.385
20.	*Paraserianthes falcataria* (L.) Nielsen.	120	100	21	4.8	0.91	0.27	5.98	0.21
21.	*Planchonela obovata* (R.Br.) Pierre	9	29	2	0.27	0.24	0.57	1.09	0.023
22.	*Protium javanicum* Burm. f.	1	4	1	0.04	0.04	0.27	0.35	0.005
23.	*Sandoricum koetjape* (Burm. f.) Merr.	8	18	2	0.32	0.16	0.54	1.03	0.027
24.	*Schleichera oleosa* (Lour.) Oken.	112	272	11	4.45	2.46	3.03	9.94	0.206
25.	*Sterculia foetida* L..	1	15	1	0.04	0.14	0.27	0.45	0.005
26.	*Tetrameles nudiflora* R. Br.	30	84	6	1.15	0.75	1.68	3.57	0.08
27.	*Toona sureni* (Blume) Merr.	302	523	20	12.08	4.75	5.43	22.26	0.368
28.	*Trema orientalis* Linn. Blume	2	25	1	0.08	0.23	0.27	0.58	0.008

D: density; C: coverage; F: Frequency; RC: Relative coverage; RD: relative density; RF: relative frequency; IIV: important index value; DI = diversity index *) trees

Table 2: Cyanide and mercury contents in above-ground parts of three species having F value of more than 10.

No	Species	F value *)	Cyanide (mg/kg)	Hg (mg/kg)	Energy (Kcal/g)
1	*Aglaia odorata* Lour.	41	0,55	0.73	2.51
2	*Aquilaria malaccensis* Lam.	17	2,91	1.79	2.23
3	*Dracontomelon dao* (Blanco) Merr.	58	4,62	5.13	2,48
4	*Duabanga moluccana* Blum.	18	9,69	7.30	3,50
5	*Erythrina orientalis* L.	10	15,70	6.76	3,01
6	*Eugenia subglauca* Koord. & Valeton.	14	3,35	4.50	2,60
7	*Kleinhovia hospita* L.	32	1,20	4.70	2.44
8	*Lagerstromeia speciosa* (L.) Pers.	11	1,05	0.63	2.38
9	*Melochia umbellata* (Houtt.) Stapf.	41	4,72	2.55	2.36
10	*Paraserianthes falcataria* (L.) Nielsen.	21	6,66	11.65	3.04
11	*Schleichera oleosa* (Lour.) Oken.	11	1,54	4.93	2.59
12	*Toona sureni* (Blume) Merr.	20	3,13	2.15	2.32

*) see Table 1

Phytostabilization potential of Duabanga moluccana, Paraserianthes falcataria and Erythrina orientalis:

Results of plant height measurements showed that the addition of compost into tailings improved growth of *Duabanga moluccana*, *Erythrina orientalis* and *Paraserianthes falcataria* (Fig. 1). Growth of plants on media with no addition of compost was significantly reduced compared to that of grown on uncontaminated soils.

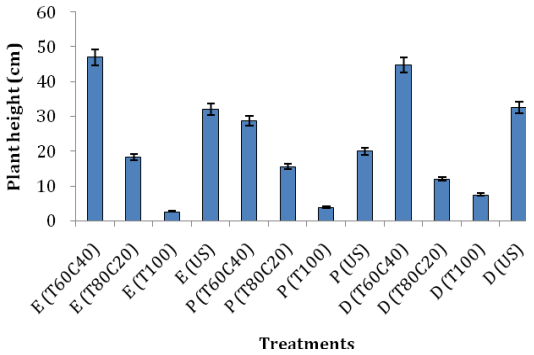

Fig. 1: Plant height of *Duabanga moluccana*, *Erythrina sp* and *Paraserianthes falcataria* grown on various mixtures of tailing and compost for 60 days. E = *Erythrina* orientalis, D = *Duabanga moluccana*, P = *Paraserianthes falcataria*; T60C20 = 60% tailing + 40% compost; T80C20 = 80% tailing + 20% compost;T100 = 100% tailing, and US = tailing uncontaminated soil.

Shoot and shoot dry weights of plants grown on media of tailing-compost mixtures were in line with the plant growth (Fig.2). This indicates that in order to grow well on media contaminated with gold cyanidation tailing, the three plant species need additional organic matter, such as compost used in this study. The optimum addition of compost was 20% by weight to reach growth that were nearly similar to that grown on uncontaminated soils. This study also showed that plants grown on tailings containing Pb and Zn was inhibited when no addition organic materials was added. This was in line with previous studied conducted by Yang *et al*. [24] and Shu *et al*. [19]. Although this study did not elucidate mechanisms involved, in general, addition of organic matter on cyanidation tailings improved water holding capacity, cation exchange capacity, and structure of the tailings through aggregate formation [25,17,9]. In addition, the added compost can stabilize metal to reduce their bioavailability [20].

Accumulation of metals in plant shoots were measured for tolerance and phytostabilization potentials of the tree species. As indicated earlier concentration of Pb in the tailings was relatively high (530 mg/kg), but Pb accumulated in the plant shoots and roots were very small (less than 60 mg/kg). Pattenrs of metals accumulation shoots and roots of *Duabanga moluccana*, *Paraserianthes falcataria* and *Erythrina orientalis* for *t*reatments D (T60C40), D (T80CK20), P (T60C40), P (T80C20), E (T60C40) and E (T80C20) were in the order of Zn > Pb > Cu > Cd (Fig.3). This indicates that accumulation of metals in plant tissues is a plant specific, especially for Zn that was accumulated greater than other metals (Pb, Cu and Cd). In addition, the amount of metals accumulated in plant tissues increased with increasing rate of the added compost. Accumulated metals ratio shoot / root on three plant species showed that all plants have metals shoot / root ratios of more than one (Fig. 4). This indicates that all plant

species tested are suitable for phytostabilization. A plant species can be considered potential for phytostabilzation if shoot : root ratio =(total element concentration in shoot tissue : total element concentration in the root tissue), are less than one [3,26]. The difference in the ratio of metals shoot / root on all plants showed differences in the effectiveness of each type of plant in transporting metals from the root system of the shoot (as a place of accumulation) [18].

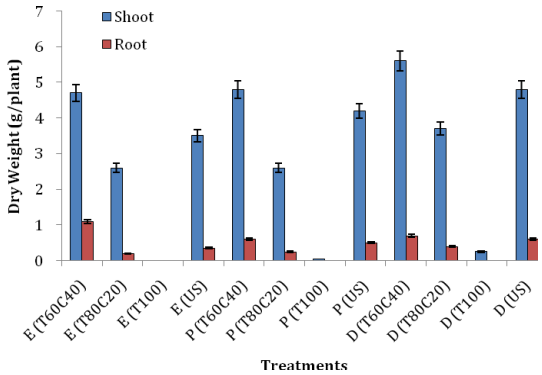

Fig. 2: Dry weight of shoot and root of *Duabanga moluccana*, *Erythrina sp* and *Paraserianthes falcataria* grown on various mixtures of tailing and compost for 60 days. Treatments are similar to those presented in Fig.1.

The concentration of Pb in plant tissues was lower than that of tailings (530 mg/kg). The highest concentration was found in the shoot *Duabanga moluccana* (29 mg/kg), and the lowest (1.8 mg/kg) was in the shoot of *Paraserianthes falcataria* grown in uncontaminated soils. The concentrations of Pb in the three plant species grown in all media were all lower than the maximum value of Pb that can be tolerated in forage, i.e. 30 mg / kg [14]. Although in most plant species, the concentration of Pb in the leaves of plants ranged from 5 to 10 mg / kg [7], the concentration of Pb in the shoots of three plants grown on tailings media did not affect plant growth. This suggests that the three types of plants were tolerant to Pb. The concentration of Zn in the plant shoots of all treatments varied from 20 mg/kg in the shoot of *Paraserianthes falcataria* grown on uncontaminated soils to 390 mg / kg in the shoot of *Duabanga moluccana* grown on a mixture of 80% tailings and 20% compost, while concentration of Zn in the tailings is 3760 mg / kg. Based on the maximum limit of Zn concentration in forage of 500mg/kg NRC, 1980), shoots of the three plant species will not harm the animal, if they should be used as animal feed. Plants grown on uncontaminated soils contains Cu 7 mg/kg on average, whereas the highest Cu concentration (27 mg / kg) was observed in the shoot of *Dubanga moluccana* planted on a mixture of 80% tailings and 20% compost. Nevertheless, this highest Cu concentration is in the normal range of 5-30 mg / kg (Kabata-Pendias and Pendias. 1992), and below the maximum tolerance concentration of Cu (100mg/kg) for animal feed NRC, 1980). The content of Cd in all treatments showed no significant differences, ranging from 0.1 mg / kg to 0.7 mg / kg, while the Cd content in the tailings is 4 mg / kg. The limit of Cd content in animal feed depends on human food waste NRC, 1980). Organic residues containing Cd of more than 0.5 mg / kg Cd in the short term is not expected to be harmful to the health of animals and humans NRC, 1980).

Phytoremediation of soils contaminated with gold cyanidation tailings containing heavy metals can focused on metal phytoextraction, or metal phytostabilization. In phytostabilization, accumulation of metals within the plant shoots is undesirable because it may be used as animal feed. Therefore, information on metal accumulation in plant tissues is very important because biomass *Paraserianthes falcataria* and *Erythrina orientalis* can be used as animal feed. Based on the analysis of the content of metals in the plant shoots and roots, indicate that *Duabanga moluccana*, *Paraserianthes falcataria* and *Erythrina orientalis* can be considered as plants that are resistant to metals or as candidates for phytostabilization strategy. Several studies of heavy metal tolerant plants had been carried out in Indonesia. Supriyanto [21] who tested to grow *Duabanga moluccana*, and *Paraserianthes falcataria* on gold mine tailings media indicated that the two plants were able to grow well with few additional inputs such as compost. *Paraserianthes falcataria* and *Erythrina orientalis* for remediation of heavy metal contaminated soils provide additional benefits as the two legume trees can supply substantial amount nitrogen through nitrogen fixation [6].

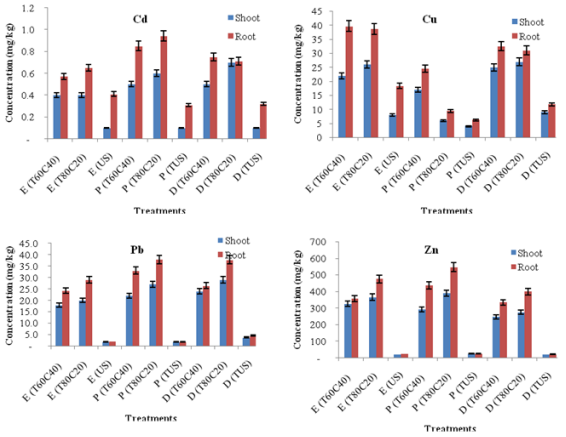

Fig. 3: Concentrations of Cd, Cu, Pb and Zn in shoots of *Duabanga moluccana*, *Erythrina orientalis* and *Paraserianthes falcataria* grown on various mixtures of tailing and compost for 60 days. Treatments are similar to those presented in Figure 1.

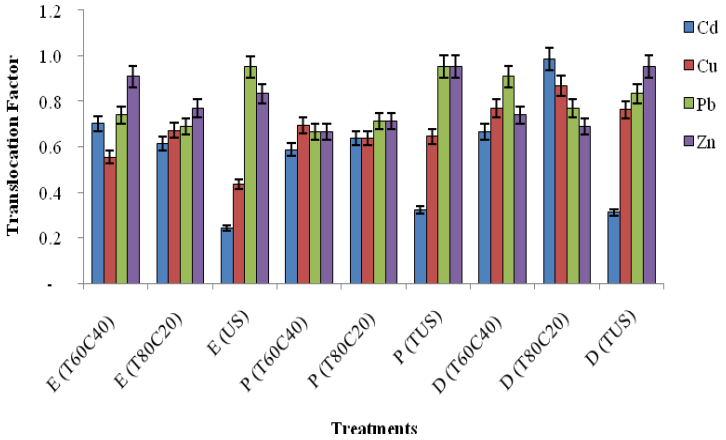

Fig. 4: Ratio of metal concentration shoot to metal concentration in root (translocation factor) of *Duabanga moluccana*, *Erythrina orientalis* and *Paraserianthes falcataria* grown on various mixtures of tailing and compost for 60 days. Treatments are similar to those presented in Figure 1.

Conclusion:

In areas contaminated by gold cyanidation tailings of Sekotong District of West Lombok there were at least 28 tree species that have long adapted and survived under extreme conditions (high metal concentrations). Amongst them, three plant species (*Duabanga moluccana*, *Paraserianthes falcataria*, and *Erythrina orientalis*) are candidates for phytostabilization strategy based on their tolerant to heavy metals.

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