Heavy metal concentration in soil and accumulation in selected plant species: A case study of Tampakan, South Cotabato

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ABSTRACT

Natural processes bring heavy metals and further escalated by a long history of addition from industrial development. The soil serves as a sink and can accumulate different types of minerals and nutrients, including metals and metalloids that are harmful even at lower concentrations. Plants' ability to sequester metals and mobilize into various plant parts are significant in the bioremediation process. This study used quantitative and exploratory research design, which involves the collection of soil and plant samples subjected to laboratory test to determine the heavy metal concentration in Pulabato, Tampakan, South Cotabato in terms of arsenic (As), copper (Cu), lead (Pb), cadmium (Cd) and mercury (Hg). Analysis of variance (ANOVA), correlation, simple and multiple linear regressions are employed to determine significant differences, relationships, and influences. Results show significant differences in plant heavy metal concentration across low, mid, and high elevations for metals Cu, Pb, and Hg. Pb concentration in soil shows a significant relationship with Cu in plants. Also, Pb concentration shows a positive correlation and Hg with a negative correlation for Pb concentration in the plant. Moreover, Hg's presence in plants shows a negative correlation in Pb for soil and a positive relationship with Hg. Simple linear regression statistics show that Pb significantly influences Cu, Pb in the plant by Pb in soil, and Pb in the plant with Hg in soil. Multiple regression statistics show a significant Pb's influence in the soil to Pb in the plant but not with Hg. Lastly, As and Pb significantly influences Hg concentration in plants.

Keywords: *heavy metals, accumulation, plants, soil, Tampakan, Philippines.*

INTRODUCTION

Heavy metals are toxic materials present in the environment which pose severe threats to humans and the environment due to its persistence and non-biodegradable nature (Nagajyoti et al.,2010) and the entry of heavy metals in the natural environment is attributed to human and natural sources (Ali et al.,2013). The contamination of soil media becomes a global concern (Zhang et al.,2020; Li et al.,2020). Plants can accumulate metals present in the soil (Nouri et al.,2009). Plants that grow in the heavy metal contaminated environment how metals and other toxicants enter the food chain (Buruka et al.,1997). In high mountain areas, the differences in elevation from a marked climate determine the vegetation distribution, and soil development affects the deposition of trace metals (Bing et al.,2016).

Tampakan is a second class municipality in the province of South Cotabato, lies 6°27′N 124°56′E, surrounded by hills and rolling terrain with tropical rainforest climate, and an estimated elevation of 134.8 meters above sea level. It serves as the food basket of the province, making the municipality more engaged in agricultural production. The understanding and investigation of the danger of substantial metal entry in the trophic level are of considerable significance to avoid detrimental effects on human health and the environment (Morkonas et al., 2018). Plant species for bioremediation processes now become the trend as one of the most effective, efficient, and low-cost technology to rehabilitate contaminated environments (Chibuiki et al.,2014).

Moreover, the study site is identified as an area with high mineral content, such as copper and gold, the potential for mining surrounded by vast agricultural lands in the lowland areas. Thus, the investigation of possible accumulation and entry in the food chain of heavy metals from soil encourages the research to conduct the study. The objective of the study is to (1) determine the significant difference between heavy metal concentration between sampling sites, (2) identify the significant relationship between soil metal and plant metal concentrations, (3) evaluate the significant influence of soil metal concentration to plant metal concentration.

MATERIALS AND METHODS

This study utilized quantitative and exploratory research designs that involve the collection of soil and plant samples at different locations. This includes the collection of soil samples and 27 individual plant species composed of trees, shrubs, grasses, and ferns known to be the dominant species present in the study area. Purposive sampling is employed considering the limitation in the collection of samples such as inaccessibility due to rugged terrain and steep slopes.

Ten most dominant plant species were collected at every 200-meter distance from every plot, which are marks as low, mid, and high elevation (Figure 1). Samples of roots, leaves, and stems underwent washing using distilled water to eliminate dirt and subjected to air and oven- drying to remove moisture. Soil samples were air-dried and sieved. 400 mg of dried specimens of roots, stems, and leaves were stored in sealed plastics bags and 500 mg of sieved soil samples. All samples were sent to F.A.S.T. Laboratories in Cagayan de Oro City for analysis.

The Study Area

The study was conducted in the proposed copper-gold mine project in Barangay Pulabato, Tampakan, South Cotabato. The study site is located at the south-central Mindanao dividing range with moderate to severely dissected with steep to very steep topography shown in (Figure 2). Ground levels vary from approximately 550 to 1,350 mean above sea level. The range rises rapidly from the surrounding plains with an abrupt change in slope. The study area's total area covers 10 thousand hectares covered with trees, grasses, vines, shrubs, herbs, and ferns. The study site includes a 600 m plot divided into low, medium and high elevations with 200 m distance each plot. As shown in Figure 1, these were the study area's exact geographical distribution across low, mid, and high elevations. Geographic coordinates of low, middle, and high elevations were plotted using Global Positioning System (GPS) receiver.

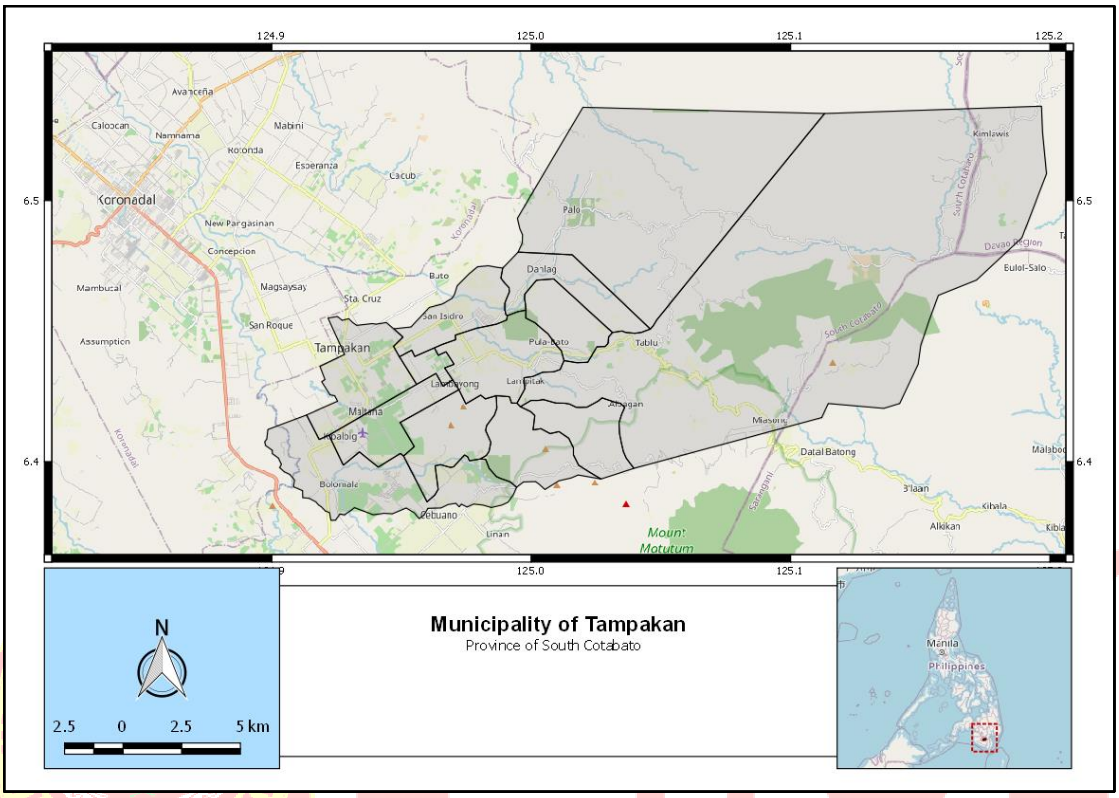

Figure 1. Map Showing the Location of the Study Site

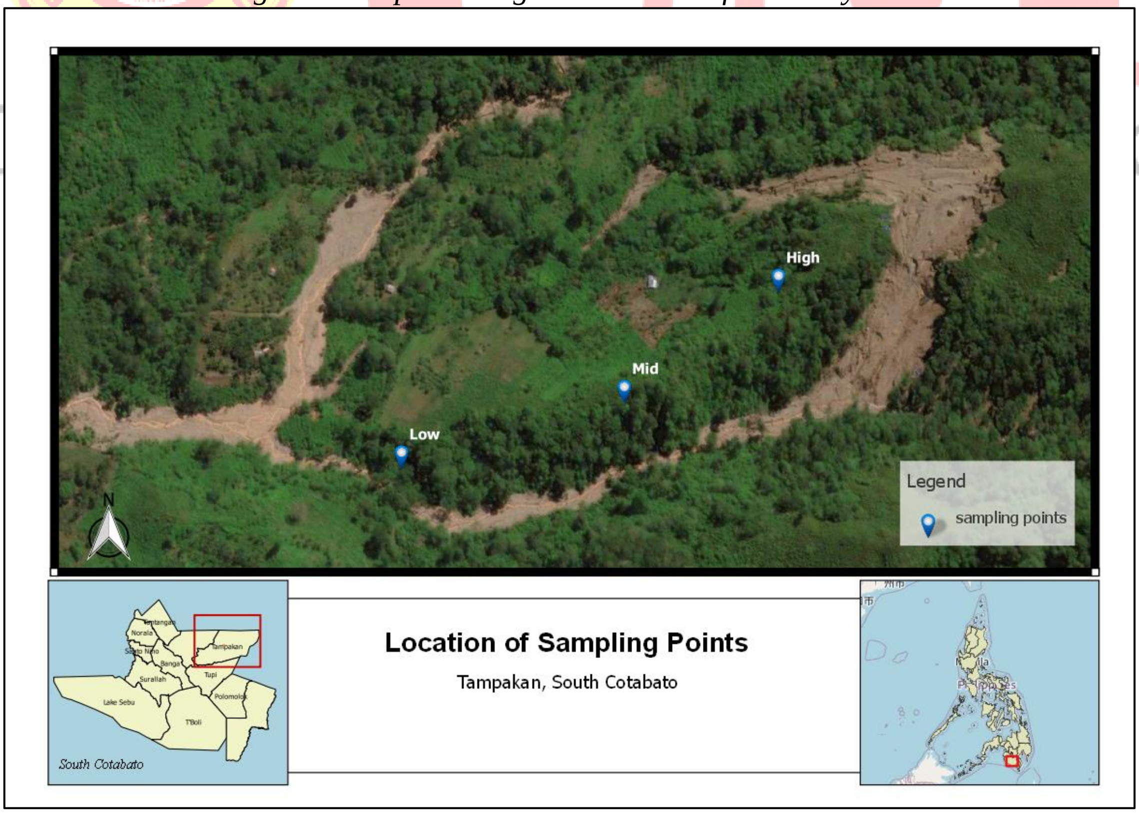

Figure 2. Map Showing the Location of the Sampling Site

Data Analysis

One-way Analysis of Variance (ANOVA) was employed to test the significant differences in heavy metal concentration among sampling sites. Moreover, a correlation was performed to identify the significant relationship between the heavy metal concentration present in the soil and the most dominant species' heavy metal accumulation. Lastly, simple linear and multiple regression were employed to determine the best fit model to describe the influence of heavy metal concentration in soil and the heavy metal concentration observed in the plant.

RESULTS AND DISCUSSION

Presented in Table 1, 2, 3, 4, and 5 are the results of the one-way ANOVA which determines the significant differences of heavy metal concentration in the soil and between and among plants sampled and analyzed for heavy metals arsenic, cadmium, copper, lead, and mercury across low, mid and high elevations.

For Arsenic (As), the soil's heavy metal concentration did not differ significantly between and among plants collected from low, mid, and high elevations based on the computed F-value of 0.60 and p-value of 0.557. The Tampakan area, particularly Pulabato, is along with the range of a volcanic arc typically influences the widespread distribution of As across the study site. Arsenic and arsenic compounds are typically considered the most abundant in the Earth's crust, and it is associated with volcanic eruptions (Mirza et al.,2014) and the As accumulation and restriction of plants are brought, the defense mechanisms are developed due to biotic and abiotic stress in the external environment (Morkunas et al.,2018). Also, concentration among plant species is dependent on the physicochemical and geological components of an area (Kazia et al.,2009), and it reduces plants reproductive capacity through losses fertility and cuts the development of reproductive organs (Singla, 2011).

Table 1. Summary of ANOVA results showing the significant difference of arsenic (As) concentration on plant among the three sampling sites

Factor	N	Mean	StDev	F-value	P-value
Low	10	60.70	169.10	0.60	0.557
Mid	10	5.05	3.00		
High	10	49.70	122.50		

*$p<0.05$

For cadmium (Cd), it also did not differ significantly across the three (3) elevations based on the computed F-value of 0.36 and p-value of 0.698 tested at a 0.05 level of significance. This means that the observed heavy metal concentration in the soil almost the same as recorded for all the plants being sampled and analyzed. Cd concentrations in plants across all elevations typically did not differ due to the detection limit of equipment that analyzed samples where it only recorded the same concentrations across low, mid, and high elevations. Moreover, Cd concentrations are influenced by geological parent materials and anthropogenic activities (Alloway et al., 1999). It is considered that the Cd concentration observed is brought by geological processes and mineralization across the Tampakan district, considering minimal industrial activities to none observed in the study site at the time of the collection of samples.

Table 2. Summary of ANOVA results showing the significant difference of cadmium (Cd) concentration on plant among the three sampling sites

Factor	N	Mean	StDev	F-value	P-value
Low	10	0.0823	0.00738	0.36	0.698
Mid	10	0.0823	0.00738		
High	10	0.0823	0.00105		

*p<0.05

Moreover, at a 0.05 level of significance, Copper (Cu) concentration observed in the soil and recorded in the plant tissues differs significantly based on the statistical analysis results using ANOVA based on the computed F-value of 3.73, p-value of 0.037. Differences in heavy metal concentration across different elevations can be attributed to metals' solubility and plants' capacity to sequester pollutants present in the soil necessary for growth and development (Chibuque & Obioro, 2014).

Table 3. Summary of ANOVA results showing the significant difference of copper (Cu) concentration on plant among the three sampling sites

Factor	N	Mean	StDev	F-value	P-value
Low	10	28.57	30.55	3.73	0.037*
Mid	10	16.86	18.56		
High	10	83.80	94.90		

*p<0.05

Table 4 shows that Pb concentration in different elevations both in soil and in the plants collected and analyzed was significantly different based on the computed F-value of 5.49 and p-value of 0.010. The translocation mechanisms are small, with slight downward movement and affected by the leaching process (Bolt et al.,1978). These differences can be attributed to the physical, biological, and plant factors govern chemical characteristics of the environment as well as the biological activities in the soil affecting bioavailability of heavy metals across sampling sites (Guala et al., 2010) and Pb accumulations and distributions, and it varies among different organs (Wang et al., 2014).

Table 4. Summary of ANOVA results showing the significant difference of lead (Pb) concentration on plant among the three sampling sites

Factor	N	Mean	StDev	F-value	P-value
Low	10	2.78	1.728	5.49	0.010*
Mid	10	3.41	0.589		
High	10	5.29	5.289		

*p<0.05

Lastly, for Mercury (Hg), the recorded or observed heavy metal concentration based on the results of the analysis in the soil across low, mid, and high elevations differ significantly with the metal concentration in the plants collected and analyzed from different heights based on the computed F-value of 17.11 and p-value less than 0.01. These differences can be credited to plant species' bioremediation potential, which is capable of sequestering elements and different uptake rates (Tangahu et al., 2011). Further, Hg uptake is governed by soil pH, particle size, cation exchange capacity, and plant factors including root surface area, exudation, and mycorrhizal transpiration rate (Sengar et al., 2010).

Table 5. Summary of ANOVA results showing the significant difference of mercury (Hg) concentration on plant among the three sampling sites

Factor	**N**	**Mean**	**StDev**	**F-value**	**P-value**
Low	10	0.131	0.0320	17.11	0.000*
Mid	10	0.143	0.0540		
High	10	0.055	0.0080		

*$p<0.05$

Table 6 below shows the correlation analysis results to determine the significant relationship between the soil metal concentration and the plant metal concentration of the most dominant species across Pulabato in Tampakan. Results show that As and Cd concentrations in plants don't have a relationship with As, Cu, Pb, and Hg concentration present in the soil with correlation values of 0.199, 0.174, 0.010, and 0.129 and -0.091, 0.069, -0.153, and 0.086 respectively. On the other hand, Cu concentration in the plant also did not establish a relationship with As, Cu, and Hg, but Pb in the soil but has a significant relationship with Cu in the plant with values of 0.321, -0.125, and -0.078 and 0.536 respectively. Heavy metals could potentially accumulate in the soil and migrate and may be absorbed by the plants through their roots and vascular systems (Sulaiman & Hamzah, 2018)

Table 6. Correlation matrix between soil and plant heavy metal concentration

Plant Heavy Metal	Soil Heavy Metal				
	As	Cd	Cu	Pb	Hg
As	0.199	-	0.147	0.010	0.129
Cd	-0.091	-	0.069	-0.153	0.086
Cu	0.321	-	-0.125	0.409*	-0.178
Pb	0.186	-	-0.338	0.536*	-0.386*
Hg	-0.492*	-	0.231	-0.672*	0.315

*$p<0.05$

Also, Pb concentration in the plant doesn't have a significant relationship with metals As and Cu with values of 0.186 and -0.338 while Pb in the plant has a significant relationship for heavy metals Pb and Hg present in the soil with values of 0.536 and -0.386, respectively. Lastly, metals Cu and Hg in the soil also don't have a significant relationship with values of 0.231 and 0.315, but for metals As and Pb, it has a significant relationship with -0.492 -0.672, respectively. The observed concentration and significant relationships explain that plant metal concentrations are typically dependent on the soil level. Plants' mineral composition is considered an outcome of its internal processes and external environment (Stein et al.,2016). The relationship of heavy metals in soils and their corresponding metal concentration in plant is governed by solubility and its forms, which turns to influence its longevity in soil solution, mobility in soils, and the uptake by plants (Cataldo & Wildung, 1978) and many plants successfully absorb contaminants in the ground for metals Pb, Cd, As and other harmful heavy metals (Sao et al., 2014).

Presented in Tables 7, 8, 9, 10 and 11 are the results of the regression analysis to determine the significant influences of heavy metal concentration on the soil and the heavy metal concentration observed in the sample tissues of the plant collected across low, mid, and high elevations in Pulabato, Tampakan.

For Cu, the results show that the soil's Pb concentration significantly influences the Cu concentration in plants based on the computed t-value of 2.37 and *p*-value of 0.025. Moreover, the regression statistics show a computed R^2 of 16.75%, which means that Pb in the soil can only predict 16.75% of the Cu concentration in plants. These heavy metal uptake and mobility along with the soil to the plant can be attributed to the transformation of its chemical forms and increased bio-availability and soil to root interface (Adriano et al., 2000).

Table 7. Regression analysis summary for predicting copper (cu) concentration in plant given lead (Pb) concentration in soil

Variable	Coefficient	S.E. Coefficient	t-value	*p*-value
Constant	-27.2	31.5	-0.86	0.395
Lead (Pb) Soil	2.043	0.861	2.37	0.025*

R^2 = 16.74%
R^2 adjusted = 13.77%
*p<0.05

On the other hand, the soil's Pb concentration significantly influences the Pb concentration in the plant with a significance value of 0.002 at 0.05 or a 5% confidence interval. Regression statistics show a computed R^2 of 28.70% influence based on the model in predicting the Pb concentration in the plant given the Pb concentration in the soil. The relationships established since plant uptake of heavy metals are correlated with plants' complex interactions with the underlying soil properties and their corresponding bio-availability and complex processes (Ernst, 1996), and Pb concentration is associated with topsoil (Ciriakova, 2009).

Table 8. Regression analysis summary for predicting lead (pb) concentration in plant given Lead (Pb) concentration in soil

Variable	**Coefficient**	**S.E. Coefficient**	**t-value**	**P-value**
Constant	0.915	0.923	0.99	0.330
Lead (Pb) Soil	0.0846	0.0252	3.36	0.002*

R^2 = 28.70%
R^2 adjusted = 26.15%
*p<0.05

For Hg, its concentration in the soil also significantly influenced the plant-based Pb concentration on the computed R^2 of 14.89% predictive influence and calculated t-ratio of -2.21 and *p*-value of 0.002. The root system acts as a barrier for Hg typically limits aerial translocation (Carbonnel et al., 2011), and usually, hg are stored in the root component, which reduces the risk of bioaccumulation and reduces health risks to humans (Peralta-Videa et al., 2009).

Table 9. Regression analysis summary for predicting lead (Pb) concentration in plant given mercury (Hg) concentration in soil

Variable	Coefficient	S.E. Coefficient	t-value	*p*-value
Constant	6.43	1.23	5.24	0.000*
Mercury (Hg) Soil	-6.63	2.88	-2.21	0.002*

R^2 = 14.89%
R^2 adjusted = 11.85%
*p<0.05

Also, the results of the multiple regression analysis show that Pb concentration in the soil significantly influences with a P-value of 0.029, while Hg does not influence Pb concentration with a p-value of 0.772. On the other hand, regressions statistics show R2 of 28.92% predictive influence. The Pb concentration in the soil shows influence wherein plants grow in a toxic environment developed phytoextraction response pattern which absorbs metals and metalloids which exhibits no damage in its external and internal structure (Barcelo and Poschenrieder, 2003)

Table 10. Regression analysis summary for predicting lead (Pb) concentration in plant given lead (Pb) and mercury (Hg) concentration in soil

Variable	Coefficient	S.E. Coefficient	t-value	*p*-value
Constant	0.09	2.98	0.03	0.976
Lead (Pb) Soil	0.0938	0.0406	2.31	0.029*
Mercury (Hg) Soil	1.24	4.24	0.29	0.772

R^2 = 28.92%
R^2 adjusted = 23.66%
*p<0.05

Lastly, regression statistics show that As and Pb concentration in the soil significantly influences the Hg concentration in the plant-based on the computed F value of -2.56 and -4.40 and p-value of 0.016 0.000, respectively. The computed R2 showed 55.90% predictive influences in predicting the Hg concentration in plants given the soil's As and Pb concentrations. Arsenic can co-exist with other metals since most As compounds strongly absorb in the soil with great attraction with sulfur (Adenji, 2004) and form other organic and inorganic arsenicals (Tangahu et al.,2011).

Table 11. Regression analysis summary for predicting mercury (Hg) concentration in plant given arsenic (As) and lead (Pb) concentration in soil

Variable	Coefficient	S.E. Coefficient	t-value	*p*-value
Constant	0.2294	0.0216	10.64	0.000*
Arsenic (As) Soil	-0.000037	0.000014	-2.56	0.016*
Lead (Pb) Soil	-0.002424	0.00055	-4.40	0.000*

R^2 = 55.90%
R^2 adjusted = 52.63%
*p<0.05

On the other hand, the presence of heavy metals observed in the plant component is influence by the bioavailability of minerals as influenced by the properties which account for metal transfer from the soil to plant (Sherenr, 2010). As show, the significant influence and Pb for Hg in the plant are attributed to the heavy metal association resulting in Hg's bioavailability for plants' uptake. Wuana and Okiemen (2011) states that changes in the form and bioavailability of the heavy metal are possible, and their soil forms and occurrences influence their mobility (Laghlimi et al., 2015).

CONCLUSION

Mining and subsequent ore-processing are always associated with extreme levels of heavy metals deposition into the environment. Moreover, the Tampakan copper-gold and the metals concentrations are naturally occurring, considering less development and industrial undertaking in the area. The uptake of heavy metals by plants and subsequent accumulation along the food chain is a potential threat to animal and human health. Physicochemical and geological also influences the absorption and accumulation of heavy metals in the environment.

The study suggests that heavy metal concentration of plant species across low, mid, and high elevations are not significant for metals As and Cd while metals Cu, Pb, and Hg differ significantly. Moreover, correlation results show that Pb in soil has a positive relationship with Cu concentration in plants. Also, Pb present in the soil has a positive relationship with Pb in the plant and a negative relationship for metals Hg. Further, arsenic in the soil also has a negative relationship with Hg in the plant and a positive relationship with heavy metal Pb. Furthermore, simple linear regression analysis shows that Pb concentration in the soil significantly influences Cu in the plant. Also, heavy metals Pb and Hg in the soil significantly influences Pb concentration in the plant. Multiple regression analysis revealed that Pb concentration in the soil significantly influences the Pb concentration in plants, but Hg does not. It can be implied that the plants in the sampling area have self-protection mechanisms that would mitigate the uptake of heavy metals from the soil (Yan et al., 2002) and both As and Pb lead concentration in the soil significantly influences Hg concentration in the plant. The accumulation of these heavy metals reduces productivity and increases health-related risks.

RECOMMENDATIONS

Further study on the soil's geochemistry, particularly in Pulabato, determines the physical, chemical, and biological factors that potentially influence the bioavailability of heavy metals for plant uptake and mobility of heavy metals. Physiological studies must be conducted to determine the physiological processes that allow and restrict metals entry in the plant systems and biophysical, geological, and chemical factors that potentially influence heavy metal sequestration. Heavy metal speciation and transformation behavior must be studied to determine the physical, chemical, and biological parameters that potentially influence its bioavailability and implication to other heavy metals present in the soil.

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