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Research Article

An Investigation of the Levels and Distribution of Selected Heavy Metals in Sediments and Plant Species within the Vicinity of Ex-Iron Mine in Bukit Besi

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An assessment of the abandoned mine impacts on the concentrations and distribution of heavy metals in surface sediments and plant species within the vicinity of an ex-iron mine in Malaysia was conducted. The sequential extraction method was used to extract anthropogenic metals in sediments. The results showed that metals in EFLE, AR, and OO fractions were higher than ambient concentrations which indicate that heavy metals have been loaded from ex-iron mining area into the surrounding aquatic environments. The metal accumulation in the four dominant plant species grown naturally within the vicinity of Bukit Besi ex-iron mining was investigated. Exceptional elevated concentrations of metal were found in plants and surface sediments. Several established criteria were applied to determine the hyperaccumulator plants. The results revealed that *Melastoma malabathricum* and *Pityrogramma calomelanos* are classified as Fe and Al hyperaccumulators, while *Scirpus triqueter*, *Melastoma malabathricum*, and *Pityrogramma calomelanos* were undoubtedly hyperaccumulator for Cd.

1. Introduction

Metal contamination in abandoned mines is a global environmental problem which various countries around the world are suffering from. This problem ranks among the most significant environmental challenges worldwide, which requires ongoing evaluation and urgent solution to overcoming this problem and its negative impacts [1]. In general, to improve the conservation of contaminated mine areas and measure the achievement goals on management of mine contamination, we need access to relevant information. Therefore, contamination of aquatic environments by heavy metal arising from both of mining processes, either underground or surface mine workings, has received growing environmental concern worldwide [2, 3]. In Malaysia, several studies revealed that mining activities have generated considerable chemical and physical changes in their surrounding aquatic environments [2, 4–6].

Bukit Besi was once an important iron mining site in the world. It was one of the world's largest iron ore producers in Southeast Asia. Several recent studies have identified the mining impacted catchments as a major source of toxic metals discharge and other pollutants in dissolved forms. In addition, these studies have discovered that the toxic elements are suspended in the material and are dispersed into surrounding river systems and floodplains in many parts of the world [7–9]. Therefore, abandoned mining sites are defined as one of the most important sources of metal pollutants [10]. In this regard, sediments consist of a complex mixture of geochemical fractions which involves various physicochemical forms of the elements. Therefore, the assessment of sediment contamination requires full knowledge of the mineral phase with which metals are associated, its physicochemical forms, and strength of binding involved [6, 11]. The sequential extraction technique has been developed to provide quality information pertaining to the physicochemical forms, processes that influence the mobilization, bioavailability, and distribution of trace metals in sediments [11, 12]. Thus, in the present study, the modified sequential extraction technique is carefully utilized to assess the distribution and levels of heavy metal in sediments.

In abandoned mining sites, high concentrations of metal are found to have significant impacts on the deterioration of the quality of the ecosystem, increasing the phytotoxicity in sediments [13]. Over time, these impacts may cause the enhancement of nutritional deficiencies, the disappearance of the natural vegetative, the decline in biological diversity, and finally posing an ecological risk to the ecosystem. The effects of these contaminants are quite varied and depend on several factors such as climatic changes, micrometeorological condition, chemical form of metal, sediments' physicochemical characteristics, and water column [14]. The hazardous metals could be transported for long distances far away from contamination sources via wind, runoff, and rainfall based on the chemical forms of the metals either in gaseous form or (particulates) by the size of the fractions of metals [15, 16].

Plants play a key role in forming a fundamental part of the trophic structure of aquatic ecosystems [9]. Plants can be considered as intermediate reservoirs of heavy metals from the sediments, water, and air and are ingested by man and animal through the food chain [17]. In abandoned mine sites, aquatic plants populations react differently to changes in the environmental factors. Recent studies have indicated that aquatic plants have a high absorption capacity of toxic metals through the roots or via precipitation within the rhizosphere and leaves during the circulation of nutrients [18–21]. Plants are capable of using metals through different ways such as complexing them in their sedentary nature, binding them into cell wall, and/or combining them to produce certain organic acid or proteins [22]. Therefore, plant species are considered as good bioindicators in the early stages of heavy metal pollution. Additionally, they can be used for monitoring the state of the aquatic ecosystem and the changes or alterations in the aquatic environments [9]. High heavy metal content in soil, water, sediments, and/or the air is found to be the most common stress factor which is faced by plant species naturally growing in abandoned mine sites. Therefore, it is imperative that plant species must adapt to different environmental conditions in order to survive. According to their adaptation strategies and heavy metals content, plant species can be classified into three main groups: metal excluders, indicators, and accumulators or hyperaccumulators [23].

Hyperaccumulator plants are widely used in phytoremediation. This is due to the fact that these plants can contain Pb, Cu, Co, Cr, and Ni >1000 μg/g or 10.000 μg/g of Fe, Mn, and Zn or Cd >50 μg/g in any aboveground tissue in their natural habitat without suffering toxic effects [23, 24]. Metal excluders can be defined as plants that can restrict translocation of heavy metals from their roots into their aboveground tissues. These species can maintain relatively low levels of metal concentrations in their shoots as compared with the elevated metals concentrations in their roots [22]. Indicator plants are plants which have the ability to accumulate the metals in their aboveground tissues; thus the metals levels in the tissues reflect the metal levels in the sediments [25, 26]. However, this type of plants dies off under continued uptake of heavy metals.

Determination of the hyperaccumulator, indicator, and exclude plant species is dependent on several criteria. A plant species can be considered as a hyperaccumulator for heavy metals if it meets one of the following four strict criteria: (1) the ratio of heavy metal concentrations of shoot to root must be higher than 1 (metal concentration in shoot/metal concentration in Root) ≥1 [25]; (2) (metal concentration in root/metal concentration in sediments or soil) > 1 [26–28]; (3) the hyperaccumulator plant must be 10–500 times greater than the same species growing in noncontaminated sites [25, 27]; and (4) plants with Pb, Cu, Co, Cr, and Ni >1000 μg/g or 10.000 μg/g of Fe, Mn, and Zn or Cd >50 μg/g in any aboveground tissue in their natural habitat without suffering toxic effects can be classified as hyperaccumulator plants [25, 27]. According to Mganga et al. [25], "a plant which has high levels of heavy metals in the roots but with shoot/root quotients less than 1 is classified as a heavy metal excluder."

Considering the above facts, the overall objectives of the present study were (1) to assess the impact of the abandoned mine on the accumulation and distribution of Pb, Al, Cd, and Fe in surface sediments and plant species that are growing on a contaminated site and (2) to identify hyperaccumulator indicator and exclude plant species using several established criteria. Therefore, the present study contributes to discovering the hyperaccumulator plants in ex-iron mine in Bukit Besi, which can be used for future studies on the management and decontamination of heavy metal-contaminated sediments using native plant species as detectors and early warning in decreasing the environmental risk posed by mining activities.

2. Materials and Methods

2.1. Study Area. Bukit Besi is an abandoned open-pit iron mine type. It was one of the world's largest iron ore producers. Bukit Besi is located about 85 km south of Kuala Terengganu, Malaysia. During 1919 a team of Japanese geologists discovered the iron ore at Bukit Besi; then in 1923 Dungun was one of the world's largest iron ore producers in Southeast Asia. In the 1960s mining activities have been stopped due to the decline in iron production. Bukit Besi covers ~2,400 ha of land. The major minerals in this area are magnetite, quartz, and goethite [2]. Recently, after the mining industry has become unproductive, the old structures, plants, and rail tracks have been left with tailings. Over time, the tailings can pose a great threat to natural reserves due to landscape changes, damage to natural drainage, pollution, and destruction of terrestrial and aquatic habitat ecosystems for decades. As the iron ore of Bukit Besi became depleted in the 1960s and 1970s, the mining has been stopped. Depending on the location of the catchments and distribution of the slag heaps, five sampling stations were selected as described in Table 1.

2.2. Samples Collection and Analysis. For sediments, surface sediments (0–5 cm depth) were collected after removing plant debris and large materials. Sediment samples were kept in precleaned plastic bags with tags and then sent to the soil laboratory in the Biology Building, at the Universiti Kebangsaan Malaysia (UKM). Five replicates of sediments

Table 1: The description and location of the sampling stations.

Station	Description	Location
1	Catchment downstream of the mining sites	4°45′2.12″N; 103°10′6.22″E
2	Leachates from the open pit mine within mine site	04°45′2.21″N; 103°10′6.52″E
3	Large catchment located directly downstream of old mine sites	03°53′59.5″N; 103°01′3.55″E
4	Stream formed by flowing ex-mine water and rainfall water	04°45′8.67″N; 103°11′2.64″E
5	Catchment located on a road in the ex-mine area	04°46′32″N; 103°10′8.89″E

were selected for each station. The complete samples were dried in room temperature until reaching a constant weight. The samples were then sieved to size of <63 μm and saved in precleaned plastic bags until analysis. The measurements of sediment characteristics (pH, grain size < 63 μm%, and percentage of organic matter) were made on the air-dried sediments. The pH was determined by the methods employed by Duddridge and Wainwright [29]. The grain-sized <63 μm% sediments were measured according to Badri and Aston [15] and the percentage organic matter was evaluated using method suggested by Walkley and Black [30]. In addition, the sequential extraction method was used as described by Badri and Aston [15].

For plant, a total of 20 plants belonging to four families, four genera, and four species were collected from different stations within the vicinity of Bukit Besi. Photographs were taken for each plants species before taking the sample. One duplicate of specimen per species was collected and placed in plastic bags with ethanol and brought to laboratory for identification. In the lab, the plant samples have been identified by horticultural botanists in botanical herbarium at Universiti Kebangsaan Malaysia; several books have been used during the identification such as [31–33]. Plant samples were washed with tap water to remove the sediment particles, followed by three times of washing, twice with distilled water and once with deionized water. The samples were dried using tissue paper and then (stems, roots, and leaves) were separated using stainless steel scissors. The separated tissues were cut into small pieces and transferred to acid-washed petri dishes and then dried to a constant weight in an oven at 70°C for three days. Finally, when samples reached the constant weight, samples were allowed to cool in the desiccators and then pulverized using laboratory mortar to produce homogeneous tissues. The metals were extracted according to the method of [34]. The description of collected plant species is given in Table 2.

2.3. Quality Assurance of Heavy Metals Analysis and Quality Control Samples. The standard reference materials (SRM) were used to evaluate efficiency of extraction methods and to validate results. In the present study, two different types of SRM were used, namely, Reference Material 8704 Buffalo River Sediment, National Institute of Standards & Technology, and LGC7162 Certificate Reference Material (strawberry leaves). In sediments samples, recoveries of the target elements were satisfactory and ranged from 102.07% to 109.86% of the certified values as shown in Table 3. In plants, recovery of the studied elements in LGC7162 Certificate Reference Material (strawberry leaves) was ranged between 86.11% and 111% from the certified value as shown in Table 4.

As a precautionary exercise to avoid the contamination during experiments, all laboratory equipment (i.e., glassware, polyethylene bottles, plastic containers, pump tubing, and plastic bags) was initially soaked in phosphate-free soap, rinsed with tap water, and then immersed in a solution of 10% nitric acid (HNO_3) for 1–3 days. It was rinsed twice with distilled water and ended with distilled deionized water, and then it was dried on a clean bench in dry room temperature. Chemical solutions which were used in the cleaning stages were also of analytical reagent grade. The laboratory equipment was permanently kept dust-free, by covering the equipment during various stages during the experimental processes. Blank samples are prepared routinely and were used to determine any contamination that may have been contributed from any sample processing steps or analytical procedure and chemical solutions.

Table 2: The scientific name, family, and collection station of the plant species.

Scientific name	Family	Collection station
Scirpus triqueter Torr	Cyperaceae	Station (5)
Melastoma malabathricum L.	Melastomataceae	Station (1)
Pityrogramma calomelanos (L.) Link	Pteridaceae	Station (4)
Blechnum orientale Linn	Blechnaceae	Station (4)

Table 3: Validation of extraction and analysis of standard references materials (sediments).

Element	Determined values	SRM values	Recovery (%)
Pb	153.1	150	102.1%
Cd	3.23	2.94	109.9%
Al	6.41	6.10	105.1%
Fe	4.15	3.97	104.5%

Table 4: Validation of extraction and analysis of standard references materials (plants).

Element	Determined values	SRM values	Recovery (%)
Pb	1.55	1.8	86.1%
Cd	0.15	0.17	88.2%
Al	111	100	111%
Fe	824.5	818	100.8%

Concentrations of heavy metal in the final solution were determined by using inductively coupled plasma mass spectrometry (ICP-MS) (model ELAN 9000 Perkin Elmer ICP-MS, USA). ICP multielement standard solution of 1000 mg/L supplied by Merck was used after dilution. In order to achieve high quality results, a calibration blank and an independent calibration verification standard were analyzed for every 20 samples to confirm the calibration status of the ICP-MS. Matrix interference (blank) was <1% for all studied elements. Metal concentrations were expressed as μg/g dry weight of sediments and plants (leaf, stem, and root).

2.4. Calculation of Hyperaccumulation Criteria. In most of the established criteria of identifying the metals accumulation plants, it is imperative to consider the metal concentrations in the aboveground biomass and the metal concentrations in the sediments or soil [24]. In addition, both of the translocation factor (TF) and the enrichment factor (EF) must be evaluated to determine that a particular plant is a metal hyperaccumulator. The enrichment factor is calculated as the ratio between the plant shoot concentrations and sediment concentrations (metal concentration in shoot/metal concentration in sediments or soil) by Branquinho et al. [28]. The translocation factor can be calculated by dividing the metal concentration in the shoot by the metal concentration in the root (metal concentration in shoot/metal concentration in Root). According to Cheraghi et al. [24], a hyperaccumulator plant should have EF or TF >1.

2.5. Statistical Analysis. SPSS version 21.0 was used to calculate the statistical analysis. The correlation between the sediments characteristics (pH, organic matter) with heavy metals concentrations at the third fraction (the oxidizable-organic fraction) was calculated using Pearson's correlation coefficients (r^2). A one-way ANOVA was used to determine significance in metal concentrations differences between the sampling stations and between the plants species and means were compared using Tukey's test.

3. Result

3.1. Validations of Analytical Methods. In sediments, recovery of all of the target elements in SRM certified standard reference materials from the sediment sample (Reference Material 8704 Buffalo River Sediment, National Institute of Standards & Technology) was ranged from 102.07% to 109.86% from the certified value as shown in Table 3. Results indicate a good recovery and tested sequential extraction is appropriate to be followed.

In plants, to evaluate the quality of extraction method, LGC7162 Certificate Reference Material (strawberry leaves) was used. A good agreement of the obtained values and the certified values was achieved. Recovery of all of the studied elements in SRM was ranged from 86.11% to 111% from the certified value as shown in Table 4.

3.2. Physical Properties of Sediments. The descriptive statistics for sediment samples basic properties (pH, OM (%), and grain size <63 μm (%)) are shown in Table 5. The sediment's pH value was extremely acidic, with values ranging between 3.45 and 3.85. On average, the grain sizes <63 μm% of studied sediments from the ex-mine catchments are determined to be only 28.94%. The percentage of organic matter (OM) content in the sediments at all the stations was low, ranging between 1.56% at station 1 and 3.12% at stations 2 and 5. The metals can be in complex form with insoluble organic compounds which therefore in effect reduces their mobility and bioavailability for aquatic organisms [35].

Table 5: The mean value of the selected sediments properties.

Station	pH	Grain size < 63 μ (%)	Organic matter (%)
1	3.45 ± 0.02	28.6 ± 0.32	1.56 ± 0.26
2	3.85 ± 0.01	26.1 ± 0.58	3.12 ± 1.18
3	3.53 ± 0.02	30.3 ± 0.91	1.90 ± 0.43
4	3.83 ± 0.024	29.6 ± 0.91	2.72 ± 0.67
5	3.67 ± 0.01	30.1 ± 0.36	3.12 ± 1.03

3.3. Heavy Metal Concentrations in Plants and Sediments. The mean concentration and percentages of Pb, Al, Cd, and Fe in the surface sediments form five different stations of the Bukit Besi are presented in Table 6. The analytical results of the present study showed that the sequence of heavy metal levels in plants was similar to those in sediments; however, some element concentrations were higher in sediments as compared with those reported in plant species. Although there are differences in exposure and uptake processes of each plant species, the mean concentrations of heavy metal in the evaluated plants species tend to decrease as the distance away from the ex-mining area along the flow direction increases. The toxic levels of Fe were found in leaves of all the analyzed plant samples.

The mean concentrations of heavy metal in different parts (leaves, stem, shoot, and root) of plants species are demonstrated in Table 7. The obtained results showed that, dependent on the most common criteria, almost all of the studied plant species were able to grow on sediments with elevated heavy metal concentrations. In the present study, four different plant species were evaluated against a number of heavy metals, namely, iron, aluminium, cadmium, and lead based metals on the several established criteria. Table 9 summarizes the results of translocation factor (TF) and enrichment factor (EF) for plant species growing around ex-iron mine. The results demonstrated that *S. triqueter*, *M. malabathricum*, *P. calomelanos*, and *B. orientale* were classified as good bioindicators plant species for Fe and Al.

4. Discussion

4.1. Heavy Metal Concentrations and Distributions in Sediments. In general, the sediment's pH value plays a major role in the controllingactivities and transfer of heavy metal in sediments. Meanwhile, the pH values of sediments at all of the sampling sites were acidic, which prevented most of the aquatic herbaceous plants from growing [17]. According to

Table 6: The mean concentration (μg/g) and percentages (%) of Pb, Al, Cd, and Fe in the surface sediments from five different stations of the Bukit Besi ex-mine catchments.

Element	(EFLE)	(AR)	(OO)	(R)	Nonresistant
			Station 1		
Fe					
Mean	1980	1423	2459	9850	
SD	122	100	57.8	97.2	37.3%
Ratio%	(12.6)	(9.06)	(15.7)	(62.7)	
Al					
Mean	52.1	55.1	1021	3034	
SD	25.4	2.71	19.3	90.3	27.1%
Ratio%	(1.25)	(1.33)	(24.5)	(72.9)	
Cd					
Mean	0.05	0.02	0.02	0.12	
SD	0.00	0.00	0.00	0.00	41.1%
Ratio%	(23.5)	(9.13)	(8.40)	(59.0)	
Pb					
Mean	0.20	0.20	0.55	13.6	
SD	0.01	0.01	0.01	2.17	6.6%
Ratio%	(1.4)	(1.4)	(3.8)	(93.4)	
			Station 2		
Fe					
Mean	3.81	251	4044	10163	
SD	3.42	4.03	40.2	628	29.7%
Ratio%	(0.03)	(1.7)	(28.0)	(70.3)	
Al					
Mean	0.56	40.0	1495	3034	
SD	0.24	0.30	14.0	80.9	33.6%
Ratio%	(0.01)	(0.88)	(32.7)	(66.4)	
Cd					
Mean	0.01	0.02	0.1	0.2	
SD	0.00	0.01	0.00	0.32	33.3%
Ratio%	(4.1)	(5.99)	(23.2)	(66.8)	
Pb					
Mean	0.08	1.7	0.92	13.8	
SD	0.00	0.04	0.90	14.4	16.3%
Ratio%	(0.48)	(10.2)	(5.59)	(83.7)	
			Station 3		
Fe					
Mean	244	653	2425	10612	
SD	47.7	3.44	115	305	23.9%
Ratio%	(1.75)	(4.68)	(17.4)	(76.2)	
Al					
Mean	20.8	77.6	2244	3034	
SD	3.08	0.79	25.1	32.5	43.6%
Ratio%	(0.39)	(1.44)	(41.7)	(56.4)	
Cd					
Mean	0.04	0.03	0.04	0.02	
SD	0.00	0.00	0.00	0.00	84.9%
Ratio%	(34.6)	(21.2)	(29.0)	(15.1)	
Pb					
Mean	0.05	0.29	0.54	14.8	
SD	0.01	0.00	0.04	0.51	5.61%
Ratio%	(0.35)	(1.84)	(3.43)	(94.4)	

Table 6: Continued.

Element	(EFLE)	(AR)	(OO)	(R)	Nonresistant
			Station 4		
Fe					
Mean	2.87	320	3511	13624	
SD	1.7	6.94	37.5	84.4	22.0%
Ratio%	(0.02)	(1.83)	(20.1)	(78.0)	
Al					
Mean	0.85	44.3	2637	3034	
SD	0.26	0.69	90.7	32.5	46.9%
Ratio%	(0.02)	(0.78)	(46.1)	(53.1)	
Cd					
Mean	0.02	0.02	0.05	0.12	
SD	0.00	0.00	0.00	0.00	44.1%
Ratio%	(8.31)	(10.1)	(25.7)	(56.0)	
Pb					
Mean	0.07	1.34	1.19	8.1	
SD	0.00	0.02	0.40	1.1	24.3%
Ratio%	(0.68)	(12.5)	(11.1)	(75.7)	
			Station 5		
Fe					
Mean	23.2	324	3503	5701	
SD	2.94	3.43	42.9	108	40.3%
Ratio%	(0.24)	(3.4)	(36.7)	(59.7)	
Al					
Mean	6.75	46.2	2170	3034	
SD	0.48	0.72	32.4	43.2	42.3%
Ratio%	(0.13)	(0.88)	(41.3)	(57.7)	
Cd					
Mean	0.01	0.01	0.05	0.2	
SD	0.00	0.01	0.05	0.01	31.4%
Ratio%	(6.07)	(4.88)	(20.5)	(68.6)	
Pb					
Mean	0.02	0.11	1.53	14.3	
SD	0.01	0.01	0.01	1.1	10.5%
Ratio%	(0.14)	(0.71)	(9.61)	(89.6)	

Peng et al. [35], a low pH increases the competition between H^+ and the dissolved metals for binding sites (OH^-, Cl^-, CO_3^{2-}, S^{2-}, and SO_4^{2-}), with low pH, dissolving metal-carbonate complexes releasing more free metal ions into the water column. In general, the low pH values are a major indictor for formation of acid mine drainage (AMD) at Bukit Besi. On average, the grain sizes <63 μm% of studied sediments from the ex-mine catchments are determined to be only 28.9%. Numerous studies reported that great quantities of metals are associated with very fine-grained particles of sediments such as clay <2 μm and <63 μm [12, 35]. This condition is due to these particles having larger surface area to volume ratio than coarse particles.

The background values of metal in sediments are not available to the public; therefore, the comparison with the average concentrations in the earth's crust [17] can be used to determine the potentially toxic or anomalous concentrations. The comparison study proved that the total heavy metals in the sediments of the Bukit Besi ex-mine were higher than the concentration of metals in natural earth crust as introduced

Table 7: Heavy metals concentrations in different plant species collected from within Bukit Besi ex-mine.

Plant	Fe (μg/g)	Al (μg/g)	Pb (μg/g)	Cd (μg/g)
S. triqueter				
Leaves	696 ± 13.2	200 ± 3.60	3.95 ± 0.01	0.28 ± 0.00
Stem	6664 ± 116	419 ± 9.04	4.00 ± 0.08	0.46 ± 0.01
Shoot	7360	619	7.96	0.74
Root	8010 ± 101	378 ± 9.58	9.79 ± 0.13	0.28 ± 0.01
Sediments	9551	5257	16.0	0.22
M. malabathricum				
Flower	35.6 ± 0.55	13.9 ± 0.11	*BDL	0.14 ± 0.00
Leaves	1388 ± 28.2	48843 ± 0.00	0.65 ± 0.01	0.07 ± 0.00
Stem	342 ± 5.64	6414 ± 46.4	2.10 ± 0.01	0.07 ± 0.00
Shoot	1766	55271	2.75	0.27
Root	224 ± 5.08	1938 ± 64.5	1.86 ± 0.04	0.07 ± 0.01
Sediments	15711	4162	14.59	0.19
P. calomelanos				
Leaves	1268 ± 40.8	527 ± 21.9	1.17 ± 0.04	0.13 ± 0.01
Stem	9873 ± 652	1493 ± 92.3	3.83 ± 0.27	0.19 ± 0.01
Shoot	11141	2019	5.00	0.31
Root	27683 ± 788	2578 ± 70.2	8.95 ± 0.16	0.24 ± 0.01
Sediments	17457	5717	10.7	0.21
B. orientale				
Leaves	823 ± 186	166 ± 39.0	1.12 ± 0.22	0.04 ± 0.01
Stem	577 ± 17.4	139 ± 6.17	2.84 ± 0.06	0.04 ± 0.00
Shoot	1400	305	3.96	0.08
Root	6425 ± 155	573 ± 10.3	5.07 ± 0.10	0.06 ± 0.00
Sediments	17457	5717	10.7	0.21

*BDL: below detection limits of ICP-MS.

by Kabata-Pendias [17]. In finer detailed explication, Al is found to be present in the earth's crust at an approximation of 8%; Fe is occurring at approximately 5%, with the average content of Cd in the earth's crust being found to be 0.1 μg/g, and Pb is reported to be approximately at 15 μg/g [17]. The present study results indicated that the highest heavy metals concentrations are found to be associated with labile fractions (EFLE, AR, and OO) at the stations within the mine site. The elevated metal concentrations associated with sediments of Bukit Besi are likely the best evidence of mining-induced influence on the sediments and aquatic environments. In addition, the presence of high content of metals in sediments is indicating that these metals are continuously dispersed downstream from the tailings by clastic movement through wind and water. In this regard, the sediments of the ex-mine areas of Bukit Besi are extremely polluted with elevated concentrations of metals due to heavily anthropogenic metal loads into the catchments from the ex-iron mines. This constitutes direct health hazards to aquatic life and human health in case of using the water of these catchments for drinking or cooking.

4.1.1. Easily, Freely, Leachable, or Exchangeable (EFLE). This fraction is used to extract easily, freely, leachable, and exchangeable ions that are weakly bound to the sediments and can be released into water by changes in the pH value or ionic competition [11, 15]. The analytical results showed that Fe was found to be the highest accumulation in the present fraction, followed by Al, Pb, and Cd. The concentrations of Fe in EFLE fraction were found to be ranging from 2.87 to 1980 μg/g in stations 4 and 1, respectively, with mean percentage of 2.93% of the total metals. In general, the analytical results proved that the presence of Fe, Al, Cd, and Pb in high concentrations in EFLE fraction indicates the occurrence of high anthropogenic loading from the mining sites into the surrounding aquatic environments. Ikenaka et al. [36] studied heavy metal levels in sediments of lake sediments in Zambia and reported that, due to the mining activities, the increase in metal pollution in Zambia is still ongoing. Yacoub et al. [11] reported that the high content of Zn, Ni, Cu, and Cd in EFLE fraction indicated a significant threat for the aquatic environment. In addition, high concentrations of Cu, Zn, and Fe in EFLE fraction were found in sediments of catchments around Sungai Lembing abandoned tin mine in a study by Ahmad and Sarah [6].

The high percentage of Fe, Al, Cd, and Pb associated with EFLE fraction would suggest that a considerable amount of Fe, Al, Cd, and Pb is becoming easily available for aquatic uptake following lowering of pH. As a summary, association of Fe, Al, Cd, and Pb with EFLE fraction is likely the best example of mining-induced influence in the Bukit Besi sediments. The results of present study were higher than those reported by [37] in which the authors studied the heavy metals concentrations in sediment of the Ngwenya iron ore mine quarry dam and reported that the metals concentrations in EFLE were 2.27 μg/g for Fe, 0.253 μg/g for Pb, and 0.318 μg/g for Cd.

4.1.2. Acid-Reducible (AR) Fraction. Hydroxylamine chloride (pH 2) (0.25 M) is usually used in AR fraction as a reagent to release metals from manganese, iron oxides, and hydroxide and possibly with carbonates too [15]. The AR fraction is used to extract metals contained in iron and manganese oxides and hydroxides which can be released under reducing conditions [11, 12]. In AR fraction, the metal concentrations were in the order of Fe, Al, Pb, and Cd. Fe concentrations were ranging from 251 to 1423 μg/g in stations 2 and 1, respectively, with a mean percentage of 4.14% of the total metals. According to Nemati et al. [38], under acidic conditions, the iron mobility enhances.

In addition, Al and Pb were also found in high concentrations; Al concentrations ranged from 44.3 to 77.6 μg/g in stations 4 and 3, respectively, and Pb concentrations ranged from 0.17 to 1.11 μg/g in stations 2 and 5, respectively. These results are in agreement with those reported by Yacoub et al. [11]. Furthermore, [39] studied the heavy metals concentrations in the former tin mining catchment and reported that Pb was dominant in the RR, followed by reducible fraction. The high abundance of Fe, Al, Pb, and Cd in AR fraction is caused by the adsorption of these metals by the Fe-Mn colloids [40, 41]. This result is at par with the findings reported by [41] that analyzed river sediment samples of Nomi River, Tokyo, Japan. The discharged effluents from inactivated iron mine may be one of the factors for the increased metals concentrations in Bukit Besi sediments. One-way ANOVA analysis showed that there are significant variations between the stations in concentrations of Fe, Al, Pb, and Cd in AR, EFLE, and OO fractions. This is most likely due to the fact that the adsorption and coprecipitation mechanisms of metals are sensitive to changes in redox potential, rendering them moderately mobile and affecting their relative concentration as well as the degree of the occurrence of AMD phenomenon and location of station from the tailings.

4.1.3. Oxidizable-Organic (OO) Fraction. According to Torres and Auleda [42], organic matter has been recognized as the main electron donor in the system; thus, the OO fraction is used to extract the oxidizable metals that are not easily released into the water [11]. In oxidizable-organic fraction, H_2O_2 has been widely used as a reagent to extract metals bounded onto organic matter due to H_2O_2 being considered as a strong oxidant to recover the organically bound fraction. From the present study, it was noted that all metals were found in high concentrations in OO fraction. Fe was the highest accumulation found in OO fraction, followed by Al, Pb, and Cd. Fe concentrations ranged from 2425 to 4044 μg/g at stations 3 and 2, respectively, which represent 23.6% of the total metals. Al concentrations were found to be high and ranged from 1021 to 2637 μg/g at stations 1 and 4, respectively, with a mean percentage of 37.3% of the total metals. These findings were in agreement with those reported by Ahmad and Sarah [6]. In addition, [39] reported that the sediments of the former tin mining catchment Bestari Jaya have been polluted by arsenic (8.8%), chromium (12.9%), copper (17.4%), lead (19.5%), zinc (14.9%), and tin (33.8%). Metals in the present fraction are not considered mobile or freely available as they are thought to be associated with stable, high molecular weight humic substances that slowly release only small amounts of metals [41].

The relationship between heavy metal concentrations at oxidizable-organic fraction and the sediment's pH value and OM% were determined. The result shows that there are significant positive linear correlations between concentrations of Fe, Cd, Al, and Pb in oxidizable-organic fraction and OM% and the pH value of the sediments. From the result, the correlation data reflect that OM% and the pH value possess a high ability to absorb Fe, Al, Pb, and Cd in the surface sediments of Bukit Besi ex-mine. The comparison of nonresistance (anthropogenic) and resistance of all studied elements in surface sediments of Bukit Besi ex-mine catchments is tabulated in Table 6. Percentage of heavy metal concentrations in the labile fractions (EFLE, AR, and OO) in sediments is shown in Figure 1. Due to the fact that fraction *R* would overshadow the bioavailable part which is of interest, it has been left out in Figure 1.

4.2. Heavy Metal Concentrations in Plant Species. The mean concentrations of heavy metal in different parts (leaves, stem, shoot, and root) of plants species are presented in Table 7 and Figure 2. The different characteristics of sediment samples determined the ability of plant species to grow. The high accumulation of metals in the surface water and sediments disabled growth of various plant species on the catchments. The results showed that all the investigated plant species have different metal-enrichment capabilities. The extent of metal accumulation in the evaluated plant species differs by species, organ, and metals. Similar observation was reported by [43]. Higher metals contents were observed to decrease in the order of root > stem > leaves of most of the plants samples, except for Fe and Al concentrations in *M. malabathricum*, of which the leaves were found to contain the highest concentrations of Fe and Al, followed by the stem, root, and flowers. The root of *P. calomelanos* had the highest Fe level of 27683±788 μg/g, while the lowest Fe value was recorded in *M. malabathricum* flower and root with the values of 35.6 ± 0.55 and 224 ± 5.08 μg/g, respectively.

The analytical results showed that the levels of Fe in the leaves of all plants species evaluated were found to have higher toxic levels of Fe in leaves, which was suggested by Kabata-Pendias [17] in Table 8. The results of the present study were highly compared by [18], which were ranged from 7.80 to 15.60 μg/g for Fe in different types of vegetables grown near sewage water area. The levels of Al found were followed by levels of Fe; the highest level of Al was 48843 ± 0.00 μg/g, which was detected in the leaves of *M. malabathricum* and in the shoots with value of 55271 μg/g, whereas the least values of Al (139 ± 6.17 and 13.9 ± 0.11 μg/g) were found in the stem of *B. orientale* and flowers of *M. malabathricum*,

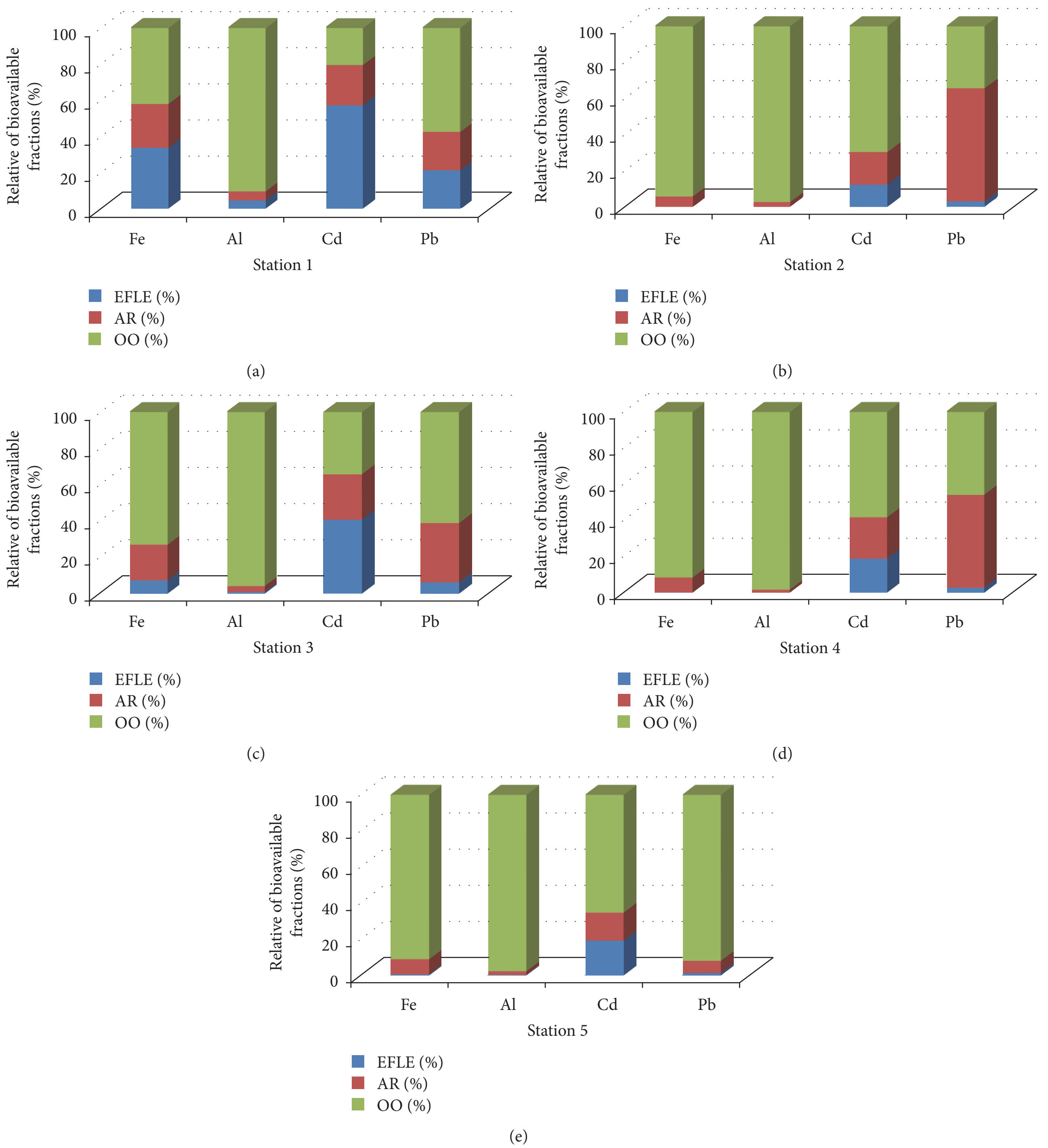

Figure 1: Percentage of heavy metal concentrations in the labile fractions (EFLE, AR, and OO) in sediments.

respectively. According to Watanabe and Osaki [44], *M. malabathricum* are woody plants which have high capacity to accumulate elevated concentrations of Al over 10.000 μg/g in their leaves as monomeric Al and Al-oxalate complexes. This phenomenon is attributed to *M. malabathricum* having high capacity for retention of Al in root symplasts rather than high Al uptake rate into the symplasts.

Reference [17] reported that Al is a common and essential element for plants and that accumulators species are plants that contain more than 1,000 μg/g of Al in their tissues. Results of the present study reported that only *M. malabathricum* and *P. calomelanos* exhibit Al concentrations in their tissues of more than the normal ranges introduced by Kabata-Pendias [17]. Regarding Pb, the highest value, 9.79 ± 0.13 μg/g, was estimated in the root of *S. triqueter*, and the lowest value, 0.65±0.01 μg/g, was found in *M. malabathricum* leaves. Therefore, Pb concentrations in all of the evaluated plant species were found to be still within the normal range as

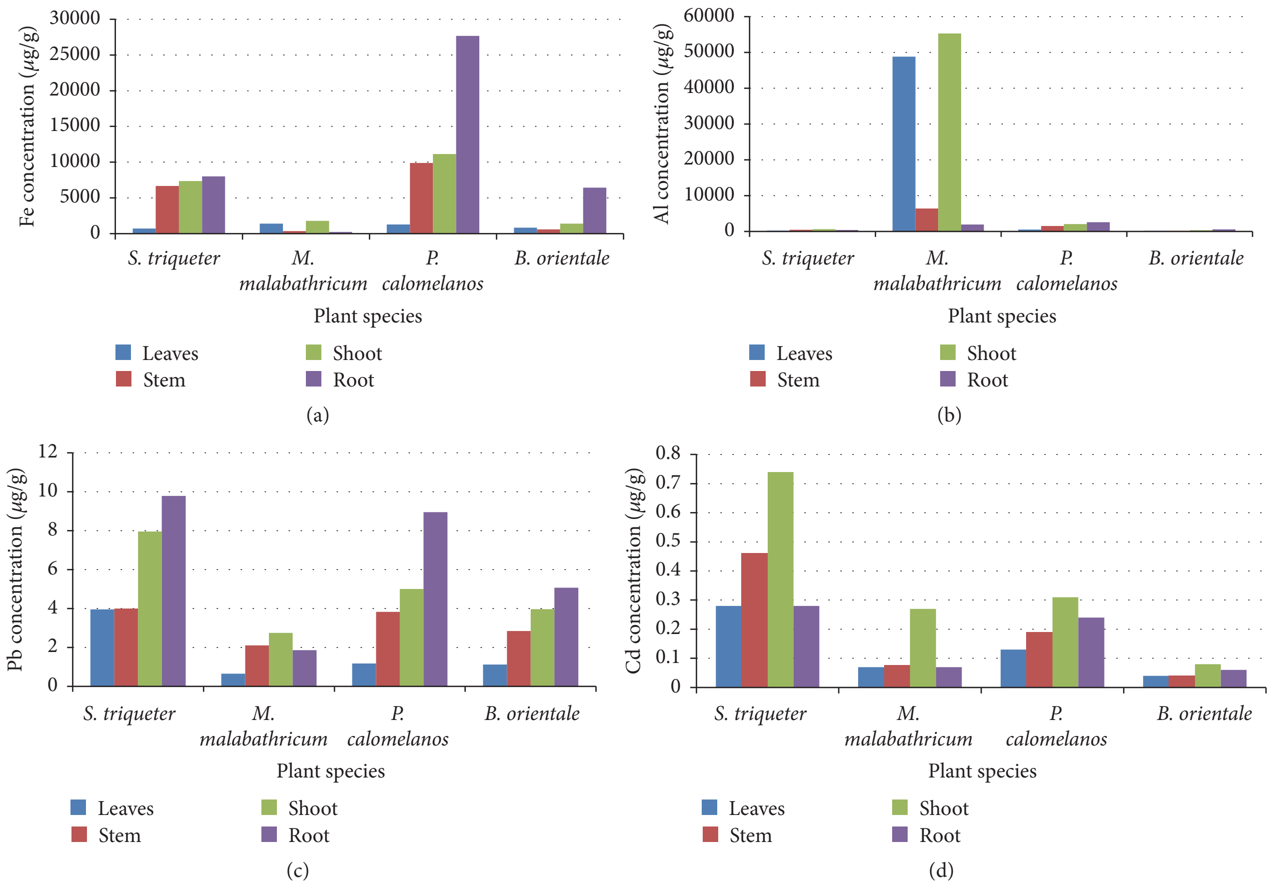

Figure 2: The mean concentrations of heavy metal in different parts (leaves, stem, shoot, and root) of plants species.

Table 8: Normal, excessive, and toxic values of metals in mature leaf tissue and vegetation generalized for various species (μg/g).

Element	Sufficient or normal values	Excessive or toxic values	Tolerable in crop plant
Cd	0.05–0.2	5–30	0.05–0.5
Pb	5–10	30–300	0.5–10
Fe	45–200	200–500	100

Table 9: Translocation factor (TF) and enrichment factor (EF) for plant species growing around ex-iron mine.

Plant Species	(Fe)		(Al)		(Pb)		(Cd)	
	(TF)	(EF)	(TF)	(EF)	(TF)	(EF)	(TF)	(EF)
S. triqueter	0.92	0.77	1.64	0.12	0.81	0.50	2.68	3.38
M. malabathricum	7.90	0.11	28.52	13.28	1.48	0.19	3.93	1.44
P. calomelanos	0.40	0.64	0.78	0.35	0.56	0.47	1.31	1.51
B. orientale	0.22	0.08	0.53	0.05	0.78	0.37	1.34	0.39

shown in Table 8. Reference [45] reported Pb concentration in root of *Calotropis procera* which was collected from the heavy traffic area site as 2 μg/g.

The highest value of Cd with the value of 0.46 ± 0.01 μg/g was determined in stem of *S. triqueter*, while the lowest value, 0.04 ± 0.00 μg/g, was recorded in the stem and leaves of *B. orientale*. According to [17, 46], "the normal concentration of Cd in leaf tissue ranges between 0.05–0.2 μg/g, and the excessive or toxic values range from 5–10 up to 30 mg/g." Thus, Cd concentrations in all of the evaluated plants were found to be within the normal range. The high concentration of Cd was between 8.85 and 18.25 μg/g in *Conyza canadensis* which was found to be growing on Mn Mine Tailings and ranged from 2.75 to 2.75 μg/g in *Poa pratensis* [10]. In addition, [22] determined the Cd concentrations in 30 species collected ore mines centers in the Gafsa-Métlaoui Basin (GMB) in Tunisia. The Cd levels were ranged from 0.11 to 82 μg/g and the highest Cd concentrations were reported in the leaves of *Anthemis Stiparum*. According to Kumar et al. [47], the aquatic plant uptakes metals either by root system or by leaves or by both ways. One-way ANOVA test showed that significant differences existed between the leaves, stem, and roots ($p < 0.05$).

As a main result, it was found that some of the plant species could grow as colonies on the contaminated water and sediments. Plant species under natural conditions can potentially uptake and accumulate some metals ions in levels exceeding the metals in the surrounding medium [22]. The results would give an indication that consuming the metals contaminated leaves by herbivores for a reasonable length of time could be a link to exposure in humans which may pose

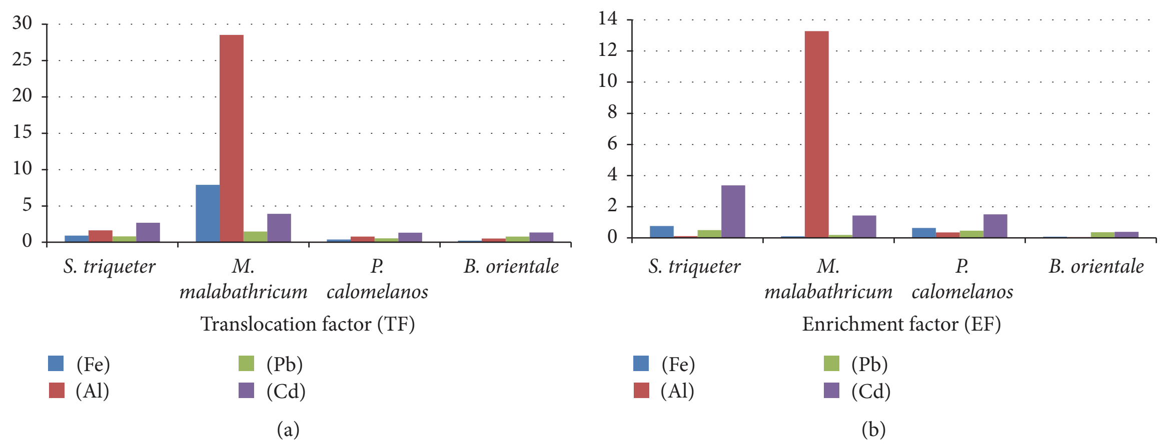

Figure 3: Translocation factor (TF) and enrichment factor (EF) for investigated plant species.

a health risk. According to [24, 48] Al, Fe, and Pb could be toxic to various plant species in concentrations > 100, 500, and 300 μg/g, respectively; thus in the present study most of plants species have heavy metals contents which are higher than the toxic levels of Al and Fe.

4.3. Identification of Tolerant and Hyperaccumulator Plants in Study Area. Considering the hyperaccumulator plants definition of [23, 24], the shoot of *M. malabathricum* and *P. calomelanos* reveals metal concentrations higher than 10,000 μg/g of Fe with value of 1766 and 11141 μg/g, respectively. In addition to this, only *M. malabathricum* had TF >1. Considering the hyperaccumulator plants definition of [25, 27], the authors proposed that the hyperaccumulator plant must contain contaminants 10–500 times greater than the same species growing in noncontaminated sites. However, in the present study, the difficulty and lack of findings derived from other locations. Thus, the normal values of metals in mature leaf tissues and vegetation generalized for various species (μg/g) introduced by Kabata-Pendias [17] were used to compare the results with the species growing in noncontaminated sites. In this regard, the concentrations of Fe in *M. malabathricum* and *P. calomelanos* were higher than those found in normal plants. *M. malabathricum* and *P. calomelanos* had values of 39.3 and 248 times greater than the normal range of Fe 45 μg/g as introduced by Kabata-Pendias [17]. According to the results, *M. malabathricum* met three criteria for Fe hyperaccumulation; therefore, it was undoubtedly Fe hyperaccumulators, while *P. calomelanos* met only two criteria for Fe hyperaccumulation.

Regarding Al, Kabata-Pendias [17] reported that Al is a common and essential element for plants. Accumulator species are plants that contain more than 1000 μg/g of Al in their tissues. According to the results, Al concentrations were found to be higher than 1000 μg/g in *M. malabathricum* and *P. calomelanos* shoot with values of 55271 and 2019 μg/g, respectively. Therefore, both of the species are considered as Al accumulators species. On the other hand, the results also show that only *M. malabathricum* had TF and EF >1 for Al. This signifies that *M. malabathricum* was undoubtedly an Al hyperaccumulator because it met three of the criteria for Al accumulators.

On the other side, *S. triqueter*, *M. malabathricum*, and *P. calomelanos* had TF and EF >1 for Cd, while *B. orientale* had only TF >1 for Cd. Therefore, *S. triqueter*, *M. malabathricum*, and *P. calomelanos* have met two of the criteria; thus these species were undoubtedly identified as hyperaccumulator for Cd. In the case of Pb, only *M. malabathricum* had TF >1 as shown in Figure 3. According to [49] hyperaccumulation of lead is particularly rare because of the low solubility characteristic of most Pb compounds. "A tolerant species is one that can grow on soil with concentrations of a particular element that are toxic to most other plants" [22, 24]. Therefore, the field study results indicated that *S. triqueter*, *M. malabathricum*, *P. calomelanos*, and *B. orientale* were the most dominant plants species, which are found to be naturally growing on extraordinarily contaminated sediments. Thus these plant species could be classified as hypertolerant to Fe, Al, Pb, and Cd metals found in the Bukit Besi ex-mine areas [10].

The results of the present study showed that *B. orientale* was identified in this study as iron and Al excluder; *S. triqueter*, *M. malabathricum*, *P. calomelanos*, and *B. orientale* were classified as good bioindicators plant species for Fe and Al. The present study also classified *S. triqueter*, *M. malabathricum*, and *P. calomelanos* as Cd indictors. It is important to note that plant species are classified into three main groups, metal excluders, indicators, and accumulators, which must be subject to several stringent standards. This is because some of the plant species are potentially classified as hyperaccumulators or excluders during their early stages of metal uptake.

5. Conclusions

The results of this investigation revealed that heavy metal levels in surface sediments of ex-mining catchments are extremely hazardous. The analytical results of the present study showed that the sequence of heavy metal levels in plants was similar to those reported in sediments; however, some element concentrations were higher in sediments as compared with those in plant species. Although there are differences in exposure and uptake processes of each plant species, in the present study, the mean concentrations of heavy metals in the evaluated plants species tend to decrease as the distance away from the ex-mining area along the flow direction increases. The toxic levels of Fe were found in leaves of all analyzed plant samples. The obtained results showed that, dependent on the most common criteria, almost all of the studied plant species were able to grow on sediments and water with elevated heavy metal concentrations. They were also able to accumulate extraordinarily high concentrations of metals such as Fe and Al. In the present study, *S. triqueter*, *M. malabathricum*, *P. calomelanos*, and *B. orientale* were classified as good bioindicators plant species for Fe and Al. Therefore, they are beneficial for carrying out phytoremediation of contaminated sediments and water and for revegetation initiatives around the Bukit Besi ex-iron mine site. However, through this study, it is ascertained that there is an urgent need for future studies on the agronomical requirements, tracing elements bioaccumulation.

Competing Interests

The authors declare that there are no competing interests regarding the publication of this paper.

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