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Assessment of heavy metal and metalloid levels and screening potential of tropical plant species for phytoremediation in Singapore

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ABSTRACT

Heavy metal or metalloid contamination is a common problem in soils of urban environments. Their introduction can be due to unpremeditated anthropogenic activities like atmospheric deposition produced by diffuse sources, construction activities and landscape maintenance. Phytoremediation is a rapidly evolving, sustainable approach to remediate the contaminated lands where metals and metalloids are highly persistent in the environment. The present work sets out to determine the level of 12 heavy metals and metalloids (As, Cd, Co, Cr, Cu, Fe, Mn, Mo, Ni, Pb, Sb and Zn) in soil and their accumulation by plant foliage found in nature parks and industrial sites in Singapore. The latter also involve the investigation of the remediation capacity of selected tropical plant species found at the sampling sites. The study is done using digestion and inductively coupled plasma-optical emission spectrometry. Eleven soil sampling sites across Singapore with 300 sampling points were selected, where soil (0–10 cm) and plant foliage samples were collected. Bioconcentration factors were determined to assess the phytoremediation potential of the collected plant species. Toxicity risk of heavy metals were assessed by comparing the target and intervention values from the soil quality guidelines by the Dutch Standard. Results of the study revealed there were regions where levels of heavy metals and metalloids were relatively high and could affect the environment and the health of flora and fauna in Singapore. Our study discovered that there were available tropical plant species (e.g., wildflowers, ferns and shrubs) which could potentially play a significant role in the remediation of contaminated lands that could open up a huge possibility of developing a sustainable and environmentally-friendly way of managing this emerging urban problem. Results showed that 12 plant species, including hyperaccumulator like *Pteris vittata, Centella asiatica*, were effective for the accumulation of heavy metals and metalloids.

1. Introduction

Urban soils are important economic, societal, and environmental assets. However, with continuous development and increasing anthropogenic activities, these ubiquitous assets are constantly transformed through mixing, importing, and exporting of material and contamination (Henderson, 2013; Morel et al., 2015). With the rapid development and expansion of industrial activities all over the world in the past century due to economic development needs, soils in urban areas may get contaminated with heavy metals and metalloids as the result of introduction of chemicals, metal mining and milling process, industrial wastes, and air-borne sources. (Wuana and Okieimen, 2011). Heavy metals and metalloids such as mercury (Hg), cadmium (Cd), lead (Pb), chromium (Cr) and arsenic (As) are categorized as non-threshold toxins which can exert toxic effects even at very low concentrations and cause significant biological toxicity effects (Rahman and Singh, 2019). There are other heavy metals and metalloids with certain biological toxicity, including zinc (Zn), copper (Cu), nickel (Ni), iron (Fe), manganese (Mn), cobalt (Co), molybdenum (Mo), antimony (Sb) (Ali et al., 2019). Therefore, remediation of heavy metals contaminations which may raise ecological risks to terrestrial plants, human health, and aquatic life, is of paramount importance. Traditional remediation practices to rectify contaminated land such as surface capping, encapsulation, landfilling,

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Abbreviation

AAS	Atomic Absorption Spectrophotometry
BCF	Bioconcentration Factor
CA	Cluster analysis
ICP-MS	Inductively coupled plasma-mass spectroscopy
ICP-OES	Inductively coupled plasma-optical emission spectrometry
KMO	Kaiser-Meyer-Olkin
PCA	Principal component analysis
PC	Principal components
SRC	Single Reactor Chamber

soil flushing, soil washing, removal and extraction techniques, solidification, chemical stability and etc. are often limited by their high capital cost and post-treatment safety issues. (Liu et al., 2018). Hence, there is a need to evaluate the potential of implementing nature-based solutions which is environmentally friendlier and more sustainable.

Phytoremediation, the process of growing plants to remedy pollutants from the environment, is a promising technology for environmental remediation (Antoniadis et al., 2017; Hinchman et al., 2000; Sarma et al., 2021). Phytoremediation is well-suited for application at very large fields where other conventional remediation technologies are not cost-effective or practical. This method is useful at sites with lower concentrations of contaminants where remediation can be carried out over long periods of time (Wang et al., 2020). Phytoremediation can also be used in conjunction with other technologies, for example chemical approaches (Nedjimi, 2021). There have been a few studies conducted elsewhere over the years on the effectiveness of phytoremediation (Ali et al., 2013; Lone et al., 2008; Yan et al., 2020); however, there are limited research studies on phytoremediation, heavy metals and metalloids contamination in soils of Singapore.

Previously, elemental concentrations of As, Cd, Cr, Cu, Ni, Pb, Zn in mangrove habitats of Singapore (Cuong et al., 2005) and Cu, Zn, Pb, Cd levels in marine sediments were measured (Goh and Chou, 1997) and the results suggested that heavy metals in marine sediment were highly dependent on sediment particle size and influenced by shipping activities. There are research studies that focused on heavy metal levels in urban community gardens, high conservation tropical forest, Singapore River and coastal sea waters in Singapore (Ang et al., 1989; ANN, 2018; Nguyen et al., 2019; Sin et al., 1991). To manage the potential risk posed by heavy metals and metalloids and to facilitate urban planning for a city state like Singapore, it is crucial to assess the concentrations of heavy metals and metalloids in the soil found in industrial estates and areas with less human activities such as nature park and natural forested areas. It is also important to examine and study the available tropical plant species growing in the various sites to better understand if these plants can be used to remediate the land in cases where the contaminants levels are higher than the target values.

The present work would provide baseline knowledge and understanding of metals and metalloids levels in soils of Singapore and identify potential tropical plant species which can accumulate heavy metals and metalloids for phytoremediation based on bioconcentration factor (BCF) values. Therefore, the aims of the study were to establish a general understanding of 12 selected heavy metal and metalloid contents in soils collected from industrial estates and natural areas at the various locations across the island, and to evaluate the accumulation abilities of the plant species present at the various sites.

2. Materials and methods

2.1. Sampling location

A total of 11 locations were selected in regions of Singapore city between latitude 1° 27′ 59.98″ to 1° 16′ 0.05″ N and longitude 103° 37′ 19.56″ E to 103° 54′ 15.12″ E, detailed information of sampling sites is shown in Fig. 1 and Table S1 (Refer to Supplementary Information). S1, S2 and S11 are nature parks and the rest of the sites are used for industrial activities.

2.2. Soil and plant leaf samples collection

Soil and plant foliage samples were collected between March 2019 and January 2020. Soil samples up to a depth of 10 cm were collected based on grid sampling method and stored in separate sealed bags for transport to the laboratory. After oven drying at 70 °C for 72 h, the soil samples were ground into powder by a grinder, and sieved through a 2 mm mesh for further analysis. The corresponding aerial parts of plants was sampled in triplicates. All plant foliage samples were washed and cleaned using deionised water, oven dried at 70 °C for 72 h followed by grinding into powder with mortar and pestle and sieved to less than 2 mm for further analysis.

2.3. Heavy metal and metalloid analysis

Inductively coupled plasma-optical emission spectrometry (ICP-OES) was used in this study to determine the concentration of As, Cd, Co, Cr, Cu, Fe, Mn, Mo, Ni, Pb, Sb and Zn in soil and plants. Microwave digestion on soil and foliage samples was carried out using an UltraWAVE™ microwave oven (Milestone Microwave Laboratory Systems, Germany) in a Single Reactor Chamber (SRC). The microwave-assisted acid digestion method, commonly used in these types of studies, was used to extract elements from soil and plant samples (Bettinelli et al., 2000; Melaku et al., 2005). Three replicates were performed for each sample. The contents of multiple elements were determined using an Optima 8300 (PerkinElmer, USA) ICP-OES. The standard calibration solution was prepared using multiple heavy metals and metalloids standard solution (PerkinElmer, USA) in 10% nitric acid. The calibration curves obtained for all the tests had correlation coefficients of 0.99 or greater.

Heavy metal metalloids levels were compared with the Dutch Pollution Standard (target and intervention values) for assessment of soil contamination degree. When the concentration of heavy metal or metalloid is below the target value, the soil is considered unpolluted. The soil remediation intervention values indicate when the functional properties of the soil for humans, plants and animals is seriously impaired or threatened (VROM, 2000).

2.4. Heavy metal and metalloid accumulating capacities

Phytoremediation efficiency of heavy metals and metalloids of various plant species was assessed using the bioconcentration factor (BCF) value. This is the ratio of heavy metal and metalloid contents in foliage to that in the soil. If the BCF value $\leq$ 1.00, it indicates that the plant can only absorb but not accumulate metal or metalloid. The plant has potential to accumulate metal or metalloid if the BCF value $>$ 1.00 (Olguín and Sánchez-Galván, 2012). BCF is calculated using the following formula:

$$BCF = [C]_p/[C]_s$$

Where $[C]_p$ is the concentration of heavy metal or metalloid in plant foliage and $[C]_s$ is the concentration of heavy metal or metalloid in soil.

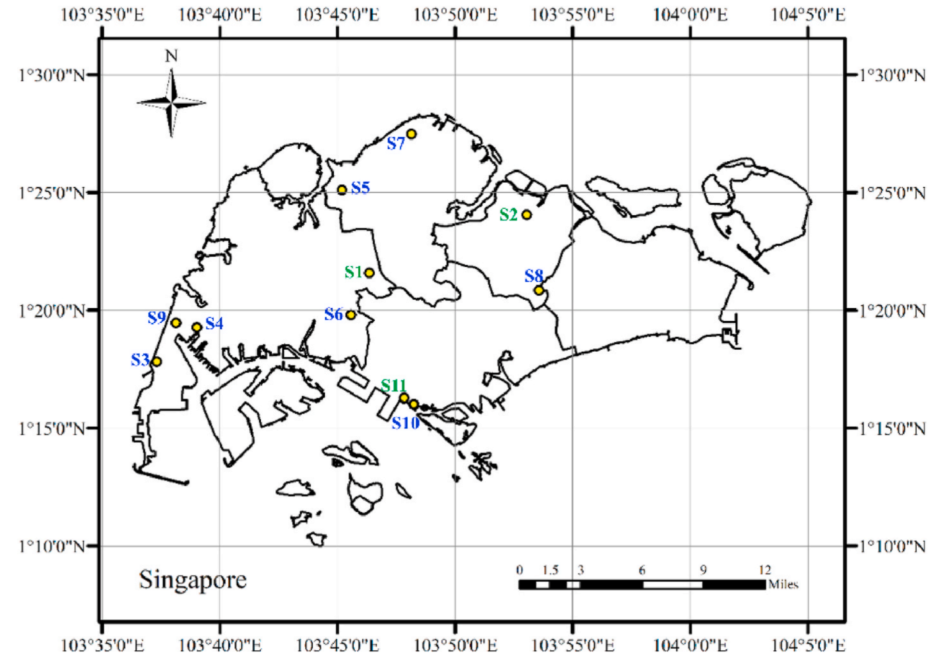

Fig. 1. Sampling locations of 11 selected sites (S1–S11) in Singapore (the map was drawn by ArcMap).

2.5. Statistical analysis

Statistical analysis was conducted using SPSS (IBM, USA). To identify the correlative relationship among heavy metals and metalloids, Pearson's correlation coefficient analysis was performed. Principal component analysis (PCA) was conducted with quartimax rotation to extract the important information from the data, to represent it as a set of new orthogonal variables called principal components (PCs), and to display the pattern of similarity of the observations and variables as points in biplots (Abdi and Williams, 2010). Cluster analysis (CA) is another commonly used multivariate statistical method in heavy metal contents analysis. CA classifies a set of cases into clusters which share common characteristics (Astel et al., 2007). In the present work, the squared Euclidean distance was used to measure similarity among clusters and Ward's method was applied as the agglomeration technique.

3. Results and discussion

3.1. Elemental concentrations in soil and plant foliage

Elemental concentrations of the 12 heavy metals and metalloids in surface soil and plant foliage from different locations were analyzed using ICP-OES are summarized in Table 1.

For environmental baseline study and environmental site assessment, Dutch standard is one of the reference standards cited within the 'Singapore Standard 593 : 2013: Code of Practice for Pollution Control' (SS 593, 2014) which is used by government agencies like JTC (JTC Corporation), NEA (National Environment Agency) and SLA (Singapore Land Authority). The target value implies that contamination is present in soil and further investigation is required, while the intervention value implies there is serious contamination of that particular metal/metalloid and cleanup is required to reduce the soil metal and metalloid concentrations to below the target value (Radomirović et al., 2020). Using the Dutch Standard as a standard for comparison (Target, 2000; Vodyanitskii, 2016), the results showed that As, Cd, Mo, Pb, Zn, Sb, Cu concentrations in some sites were higher than target values. Detailed values of each analyte were highlighted in Table 1. A summary of descriptive statistics of soil heavy metals and metalloids is shown in Table S2 and Figure S1 in Supplementary Information. It was observed that Co, Cr, Ni levels were below the target values (9.0 mg/kg, 100.0 mg/kg and 35.0 mg/kg respectively) in soil from all sampling sites. Soil Co level ranged from (0.67 $\pm$ 0.35) mg/kg (S1) to (7.78 $\pm$ 38.59) mg/kg (S7); Cr level ranged from to (8.69 $\pm$ 9.23) mg/kg (S1) to (89.4 $\pm$ 96.0) mg/kg (S5); Ni level ranged from (4.75 $\pm$ 2.48) mg/kg (S1) to (22.3 $\pm$ 34.9) mg/kg (S5).

Although Fe and Mn are not included in Dutch Standard, their values were compared to other studies. Results suggested that the mean concentration of Mn in soil ranged from (76.2 $\pm$ 133.3) mg/kg (S1) to (509.2 $\pm$ 578.7) mg/kg (S5), and this is within the range of natural background concentrations of Mn in soil, which is 0.5 mg/kg to 5000 mg/kg (Hernandez-Soriano et al., 2012). Fe levels in soil had an average value of 19,355.66 mg/kg and they ranged from (13,860.11 $\pm$ 667.10) mg/kg (S1) to (231,143 $\pm$ 16,473) mg/kg (S5). It is reported that typical Fe levels in soils range from 0.2% to 55% (20,000 mg/kg to 550,000 mg/kg) (Bodek et al., 1988), showing that Fe concentration was in a common range.

All heavy metals and metalloids levels from S11 were less than the target values, detailed values are shown in Table 1, indicating that the nature park in the central region is relatively pollution free. Incidentally, S6, showed the same trend as the S11. On the other hand, higher concentration of Cd in S1 and S2 might be due to geologic activity or ecological recycles, for instance, the flux of Cd in the atmosphere may have entered the soil through natural precipitation and rain precipitation (Cullen and Maldonado, 2013; Friedland, 1990).

In the industrial sampling sites (S3 and S9), Cd, Cu, Mo, Sb and Zn concentrations were slightly greater than target values. As, Cd, Cu, Mo and Sb levels in S4, As, Cd, Cu, Sb and Zn levels in S7, Cd, Cu and Zn levels in S8, As and Cd levels in S10 were higher than target values. It is notable that S5 contained the highest Cd, Co, Cr, Cu, Fe, Mn, Mo, Ni and Pb among the surveyed sites. Detail values are as follows: Cd - 1.77 mg/kg, Cr - 89 mg/kg, Cu - 258 mg/kg, Fe - 23,114 mg/kg, Mn - 509 mg/kg, Mo - 11.7 mg/kg, Ni - 22.3 mg/kg and Pb - 242 mg/kg. Especially for Cu and Sb. They were higher than the intervention value according to the Dutch Standard, while the highest Sb among surveyed sites appeared to be in the industrial site at the western region (S4). This could be contributed by the heavy traffic and industrial activities in urban environment (Yan et al., 2019). Overall, the results suggested that the industrial site S5 where concentrated industries such as waste management, construction at the northern part of Singapore contributed

Table 1
Heavy metal and metalloids concentrations in surface soil and plant foliage of collected locations in Singapore.

Sample site	Elemental concentrations (mean ± SD) in soil (mg/kg dry wt.) and plant foliage (mg/kg dry wt.)											
	As	Cd	Co	Cr	Cu	Fe	Mn	Mo	Ni	Pb	Sb	Zn
Site 1 Soil	**34.650 ± 42.865**	**1.655 ± 1.649**	0.856 ± 1.212	8.690 ± 9.226	14.236 ± 14.103	13860.055 ± 666.980	76.211 ± 133.280	**4.808 ± 7.512**	4.749 ± 2.478	18.140 ± 12.310	2.033 ± 1.175	71.466 ± 45.661
Plant foliage	1.117 ± 1.594	0.415 ± 0.232	0.500 ± 0.365	2.123 ± 4.155	16.257 ± 8.530	115.475 ± 72.050	103.607 ± 130.345	1.765 ± 0.865	3.698 ± 3.848	2.464 ± 12.520	0.027 ± 1.106	31.461 ± 21.875
Site 2 Soil	16.134 ± 10.699	**1.649 ± 3.699**	0.668 ± 0.345	20.709 ± 22.048	5.905 ± 40.460	15861.600 ± 9579.559	133.241 ± 96.936	1.059 ± 0.877	6.219 ± 6.591	37.094 ± 26.977	2.312 ± 1.649	44.508 ± 26.459
Plant foliage	3.970 ± 2.112	0.221 ± 0.117	0.227 ± 0.099	0.665 ± 0.640	12.095 ± 3.973	166.571 ± 144.395	27.927 ± 14.069	2.515 ± 1.183	0.338 ± 1.266	0.013 ± 0.050	0.034 ± 0.104	34.483 ± 20.661
Site 3 Soil	24.040 ± 9.367	**1.479 ± 0.445**	3.672 ± 0.952	36.188 ± 14.129	**92.357 ± 74.252**	19338.991 ± 3345.693	221.478 ± 58.245	**5.957 ± 2.077**	16.720 ± 5.385	46.963 ± 17.755	**7.103 ± 2.579**	**708.298 ± 322.256**
Plant foliage	2.597 ± 0.473	0.507 ± 0.177	0.588 ± 0.254	3.550 ± 1.645	28.625 ± 5.131	1154.097 ± 483.966	31.484 ± 6.882	3.614 ± 1.179	2.197 ± 2.003	4.949 ± 2.864	1.677 ± 0.489	336.258 ± 99.107
Site 4 Soil	**46.187 ± 12.763**	**1.629 ± 2.828**	5.484 ± 5.726	31.492 ± 29.714	**119.036 ± 208.146**	18656.300 ± 11530.670	178.981 ± 207.022	**8.953 ± 9.959**	19.013 ± 19.864	36.832 ± 48.882	**36.832 ± 48.882**	80.180 ± 324.060
Plant foliage	2.578 ± 1.330	0.024 ± 0.047	0.178 ± 0.072	1.559 ± 0.578	12.925 ± 5.614	490.171 ± 247.869	30.518 ± 7.966	5.915 ± 5.965	1.004 ± 0.646	6.653 ± 2.905	0	73.210 ± 24.923
Site 5 Soil	**33.962 ± 21.254**	**1.767 ± 0.688**	7.070 ± 24.360	89.420 ± 96.029	**258.069 ± 33.185**	23113.940 ± 16473.310	509.243 ± 578.735	**11.684 ± 13.136**	22.291 ± 34.863	**242.382 ± 1558.160**	**9.293 ± 23.486**	**406.178 ± 400.362**
Plant foliage	13.050 ± 74.624	0.517 ± 1.960	0.557 ± 0.312	6.225 ± 6.103	25.999 ± 12.567	1131.335 ± 1494.064	78.438 ± 71.125	6.377 ± 9.893	2.107 ± 1.323	3.107 ± 4.147	0.064 ± 0.158	134.379 ± 124.269
Site 6 Soil	15.447 ± 8.221	0.660 ± 0.688	3.124 ± 2.246	19.443 ± 15.239	29.532 ± 8.894	21226.063 ± 14092.900	185.524 ± 177.186	**4.158 ± 11.237**	8.937 ± 5.804	10.380 ± 14.497	0.361 ± 0.555	39.592 ± 12.617
Plant foliage	2.065 ± 2.130	0.343 ± 0.398	0.392 ± 0.235	3.220 ± 1.897	7.568 ± 4.749	593.809 ± 997.197	106.765 ± 96.403	1.640 ± 1.071	1.856 ± 1.459	0.442 ± 1.033	0.029 ± 0.071	40.213 ± 27.882
Site 7 Soil	18.220 ± 22.220	**0.900 ± 0.767**	7.775 ± 38.585	31.217 ± 29.627	**71.618 ± 76.633**	16658.293 ± 7054.399	123.021 ± 103.369	2.775 ± 2.841	10.548 ± 8.706	22.003 ± 31.184	2.474 ± 11.981	**191.728 ± 167.828**
Plant foliage	3.640 ± 8.815	2.592 ± 7.160	1.015 ± 2.338	14.774 ± 34.024	45.216 ± 145.989	6260.645 ± 14586.305	113.564 ± 186.187	2.425 ± 4.984	8.772 ± 23.858	28.199 ± 82.296	4.882 ± 18.805	402.584 ± 981.555
Site 8 Soil	**31.276 ± 24.507**	**1.618 ± 2.533**	1.530 ± 2.379	33.266 ± 30.318	**52.853 ± 107.109**	19051.862 ± 8758.958	117.337 ± 136.652	2.641 ± 3.820	16.475 ± 19.021	33.750 ± 62.198	**4.248 ± 13.931**	**208.163 ± 102.224**
Plant foliage	1.132 ± 5.250	2.916 ± 7.565	0.508 ± 0.438	3.517 ± 2.712	13.009 ± 7.288	977.625 ± 3494.809	29.931 ± 22.707	2.522 ± 1.663	2.918 ± 5.367	0.812 ± 0.929	0.235 ± 0.455	100.186 ± 94.529
Site 9 Soil	20.044 ± 6.818	**1.594 ± 0.841**	2.811 ± 1.758	46.134 ± 28.741	**86.418 ± 61.806**	20064.184 ± 7750.263	218.313 ± 99.826	**5.303 ± 3.348**	20.591 ± 13.244	53.041 ± 39.372	**5.466 ± 5.061**	**614.027 ± 480.505**
Plant foliage	0.329 ± 0.989	0.946 ± 0.985	0.692 ± 0.505	7.047 ± 6.625	30.618 ± 16.376	1682.693 ± 1582.052	40.056 ± 26.593	5.107 ± 2.880	3.762 ± 3.558	9.863 ± 10.596	0.332 ± 0.751	228.475 ± 182.512
Site 10 Soil	**42.393 ± 77.770**	**1.353 ± 1.818**	1.545 ± 1.084	32.500 ± 9.35	16.244 ± 5.756	19034.781 ± 5254.988	65.056 ± 38.682	1.335 ± 0.736	18.371 ± 15.183	72.953 ± 72.058	2.240 ± 1.396	79.224 ± 36.125
Plant foliage	1.187 ± 1.027	0.248 ± 0.398	0.464 ± 0.090	2.577 ± 2.019	6.532 ± 2.115	320.115 ± 347.969	199.821 ± 316.346	1.432 ± 1.254	3.726 ± 2.445	4.322 ± 4.310	0.040 ± 0.113	161.934 ± 143.165
Site 11 Soil	17.033 ± 8.507	0.457 ± 0.309	0.926 ± 0.478	36.744 ± 15.476	9.380 ± 20.727	20940.352 ± 7750.263	62.159 ± 47.873	1.368 ± 1.140	6.882 ± 3.858	13.645 ± 26.450	1.454 ± 0.828	52.988 ± 111.092
Plant foliage	0.811 ± 0.62	0	0.501 ± 0.064	1.199 ± 0.866	4.066 ± 2.922	119.697 ± 68.157	43.043 ± 41.094	1.227 ± 1.222	0.691 ± 0.652	0	0	28.997 ± 11.724
Target Value	29.0	0.8	9.0	100.0	36.0	–	–	3.0	35.0	85.0	3.0	140.0
Intervention Value	55.0	12.0	240.0	380.0	190.0	–	–	200.0	210.0	530.0	15.0	720.0

Heavy metals and metalloids concentration in soil which exceed target values are highlighted in Table 1.

the highest heavy metals and may have ecological risks. The concentration of investigated metals and metalloids found in soil and foliage may be correlated, and this will be discussed in multivariate analysis in the later section.

Mean concentrations of As, Cd, Co, Cr, Cu, Fe, Mn, Mo, Ni, Pb, Sb and Zn in plant foliage collected from selected 11 sites are listed in Table 1. Descriptive statistical analysis was summarized in Table S3 and Figure S2 in Supplementary Information. Foliar metals and metalloids include Co, Cr, Cu, Fe, Ni, Pb, Sb and Zn showed the highest level in S7 located on the north side of Singapore where the waste management plant and the incineration plant is located. S5 had the highest concentration of foliar As and Mo. Highest Cd and Mn in plant foliage was found in S8 and S10 respectively. Detailed description statistics analysis results are summarized in the supplementary information.

Table 2 shows the results of Pearson correlation analysis. For heavy metals and metalloids in soil, strong correlation with coefficients above 0.7 were observed between several investigated elements (Ratner, 2009). For instance, Cr concentration in soil were strongly correlated to other metals, especially Cu, Fe and Ni ($p < 0.01$), suggesting the possibility of their common presence in the region (Zhang et al., 2019). Cu concentration in soil was highly correlated with Pb and Zn ($p < 0.01$), showing that similar sources such as pesticides or fertilizers (Tariq et al., 2016). In plant foliage, Co was significantly correlated with Cr, Cu, Fe, Ni, Pb, Sb ($p < 0.01$); Cr concentrations were highly correlated with metals including Cu, Fe, Ni, Pb, Sb ($p < 0.01$); Cu level was greatly correlated with Fe, Ni, Pb, Sb ($p < 0.01$). Strong correlation also existed between Fe and Ni, Pb, Sb; Ni and Pb, Sb, Zn; Pb and Sb. The strong correlation between foliar heavy metals may also indicate their possible common origin ($p < 0.01$) (Zheng et al., 2013). It is notable that Cr was strongly correlated with Cu, Fe, Ni, and Cu was highly correlated with Pb were found both in soil and foliage.

3.2. Multivariate statistical analysis

3.2.1. Principal component analysis

PCA reduces the dimensionality of the data by identifying the principal components that represent the majority of the variance of the associated variables. In order to understand how the variables (in our case, heavy metals and metalloids concentration) which are characteristics of the sample (soil, plant), determine their association. PCA was applied to determine relationship between heavy metals or metalloids in soil and their sources (Davis et al., 2009). In the present work, PCA was conducted on the soil and plant foliage data collected at the various sampling sites. Table 3 demonstrates PCA results with component matrix post rotation of heavy metal and metalloid contents from soil and plant foliage. The Kaiser-Meyer-Olkin (KMO) measure of sampling adequacy for the above variables were found to be 0.789 and 0.892 respectively. Two principal components (PCs) with eigenvalues greater than 1 were extracted in soil and plant foliage data.

As shown in Fig. 2 (a), for soil, PC1 captures Cr, Cu, Fe, Mn, Mo, Ni, Pb, Sb and Zn, which could be explained as due to anthropogenic activities. Cu, Fe, Ni, Pb, Zn are commonly used in industrial metal products of with wide application such as pigments, paints, batteries, cosmetics (Tchounwou et al., 2012). They may be released and settle into the soil in sampling sites. Cu contents were significantly correlated with Pb and Zn, similar to the result from Pearson correlation analysis, suggesting their common sources. The high Mn level in soil may originate from industrial emissions and fossil fuel combustion. Major sources

Table 3
Results of PCA.

Metal/metalloid	Soil		Plant Foliage	
	PC1	PC2	PC1	PC2
% of Variance	47.226	10.942	58.665	10.862
As	−0.121	0.790	−0.094	0.622
Cd	0.188	0.798	0.160	0.870
Co	0.155	0.191	0.922	0.066
Cr	0.869	0.081	0.926	0.075
Cu	0.918	0.109	0.960	0.109
Fe	0.807	0.160	0.916	0.079
Mn	0.710	0.047	0.485	0.081
Mo	0.766	0.197	0.293	−0.037
Ni	0.769	0.148	0.928	0.103
Pb	0.731	−0.120	0.949	0.104
Sb	0.627	0.141	0.958	0.115
Zn	0.755	0.192	0.587	0.529

Table 2
Pearson correlation matrix heavy metals and metalloids.

Type	Variables	As	Cd	Co	Cr	Cu	Fe	Mn	Mo	Ni	Pb	Sb
Soil	As	1										
	Cd	0.313[a]	1									
	Co	−0.011	0.135[b]	1								
	Cr	0.007	0.187[a]	0.226[a]	1							
	Cu	−0.023	0.287[a]	0.184[a]	**0.743**[a]	1						
	Fe	0.049	0.263[a]	0.135[b]	**0.701**[a]	0.699[a]	1					
	Mn	0.014	0.116	0.068	0.679[a]	0.611[a]	0.541[a]	1				
	Mo	0.134[b]	0.195[a]	0.066	0.636[a]	0.671[a]	0.595[a]	0.667[a]	1			
	Ni	−0.002	0.254[a]	0.129[b]	**0.808**[a]	0.654[a]	0.597[a]	0.396[a]	0.513[a]	1		
	Pb	−0.056	0.064	0.046	0.575[a]	**0.751**[a]	0.584[a]	0.340[a]	0.441[a]	0.433[a]	1	
	Sb	−0.016	0.240[a]	0.073	0.375[a]	0.603[a]	0.474[a]	0.285[a]	0.536[a]	0.425[a]	0.438[a]	1
	Zn	−0.045	0.347[a]	0.103	0.555[a]	**0.733**[a]	0.564[a]	0.498[a]	0.540[a]	0.629[a]	0.423[a]	0.479[a]
Plant foliage	As	1										
	Cd	0.240[a]	1									
	Co	−0.014	0.195[a]	1								
	Cr	0.011	0.210[a]	**0.810**[a]	1							
	Cu	0.030	0.228[a]	**0.924**[a]	**0.867**[a]	1						
	Fe	0.032	0.201[a]	**0.809**[a]	**0.907**[a]	**0.865**[a]	1					
	Mn	−0.034	0.110	0.380[a]	0.405[a]	0.394[a]	0.385[a]	1				
	Mo	−0.015	0.064	0.229[a]	0.271[a]	0.288[a]	0.228[a]	0.034	1			
	Ni	0.004	0.234[a]	**0.841**[a]	**0.888**[a]	**0.879**[a]	**0.837**[a]	0.464[a]	0.193[a]	1		
	Pb	0.010	0.234[a]	**0.882**[a]	**0.859**[a]	**0.935**[a]	**0.867**[a]	0.405[a]	0.226[a]	**0.866**[a]	1	
	Sb	0.013	0.256[a]	**0.894**[a]	**0.860**[a]	**0.957**[a]	**0.873**[a]	0.412[a]	0.224[a]	**0.897**[a]	**0.957**[a]	1
	Zn	−0.008	0.554[a]	0.556[a]	0.531[a]	0.592[a]	0.519[a]	0.380[a]	0.153[b]	0.558[a]	0.576[a]	0.569[a]

Coefficients greater than 0.7 are in bold.
[a] Correlation is significant at the 0.01 level.
[b] Correlation is significant at the 0.01 level.

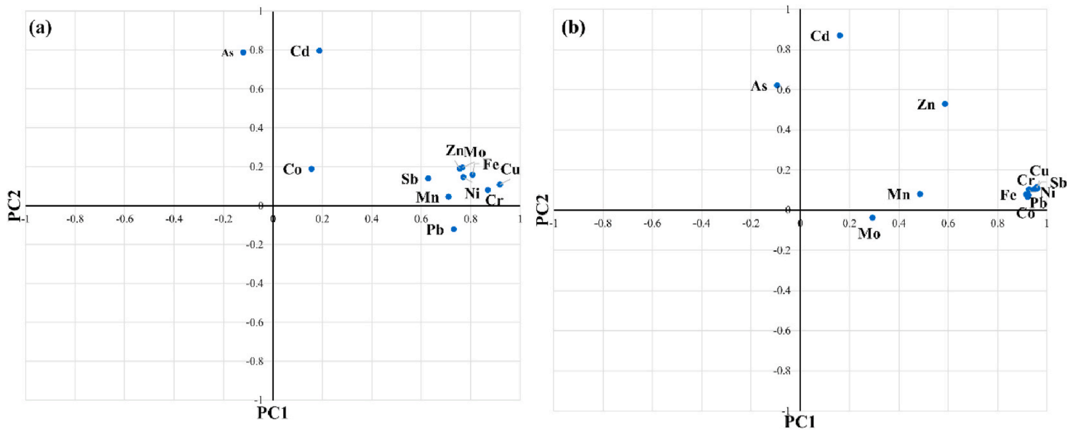

Fig. 2. Principal component analysis (PCA) loading plot for elements from (a) soil, (b) plant foliage.

of Cr include releases from electroplating processes and the disposal chemical wastes (Smith, 1995). Higher concentration of Sb might originate from vehicle fume in dense traffic areas (Ozaki et al., 2004). PC1 captured heavy metals and metalloids were mainly accumulated in western and northern industrial areas (S3, S4, S5 and S7), where there was an energy plant, an incineration plant, furniture making industries, construction, and waste management industries. On top of all these, these locations also had a high volume of traffic as well. PC2 for soil heavy metals and metalloids captures As, Cd, Co which were accumulated in natural parks (S1 and S2) could be contributed by natural sources. The distribution of As in surface soil is related to hydrothermal activity, biogeochemical processes, erosion rate (Hartley et al., 2020; Masuda, 2018; Ortiz Escobar et al., 2008). Cd is usually presents as an impurity in phosphatic rocks, and derived from animal wastes in the environment (Adriano, 2013). The content of Co in soils depends on the parent materials like rocks where Co is derived from (Ma and Hooda, 2010). S1 is located in a primary rainforest area where leaching could occur, S2 is situated near the wetland where there have some active biogeochemical activities. Therefore, PC2 can be mainly attributed to heavy metals and metalloids from natural sources.

The results of PCA analysis for heavy metals and metalloids in plant foliage determined the possible sources of tested elements in collected plant samples. In Fig. 2 (b), PC1 includes Co, Cr, Cu, Fe, Mn, Mo, Ni, Pb, Sb and Zn, and it could be an anthropogenic component (Wuana and Okieimen, 2011). Among them, Co, Cu, Fe, Mn, Mo, Ni and Zn were clustered together might indicate for plant essential metals, (Jadia and Fulekar, 2009). As and Cd are well represented by PC2, this clustered pattern implies that they come from nature. For instance, phosphate rocks, and manure are important sources for As and Cd (Chen et al., 2008; Zhang et al., 2021). As and Cd are non-essential elements for plants (Gupta et al., 2011; Leavitt et al., 1979).

3.2.2. Cluster analysis

Hierarchical cluster analysis was performed based on principal component analysis to identify metal and metalloid contents which belong to independent clusters. Heavy metals and metalloids in a cluster are followed similar pattern, but different from the others in another cluster.

In soil, the heavy metals and metalloids clustered are shown in Fig. 3 (a). There are two main clusters in the dendrogram obtained from the cluster analysis conducted using the Ward's method which makes use of the squared Euclidean distance as a similarity measure. Cluster I includes the following elements: Cr, Ni, Mn, Mo, Zn, Fe, Zn, Sb, Mn and Mo. Cluster II comprises of As, Cd, and Co. As PCA analysis results shown that PC1 captures heavy metals and metalloids (Cr, Ni, Mn, Mo, Zn, Fe, Zn, Sb, Mn and Mo) in the upper soil from anthropogenic activities, and PC2 captures elements (As, Cd, and Co) from natural sources. The interrelated association among these heavy metals and metalloids (Cluster I and Cluster II) showed same category as PC1 and PC2 of PCA. Therefore, the cluster separation of these heavy metal and metalloids suggests different origins of the elements in soils.

Cluster analysis of plant foliage heavy metals and metalloids is shown in Fig. 3 (b). There are two main clusters, cluster I contains elements Pb, Sb, Cu, Cr, Ni, Fe, Co, Zn, Mn, As, and Mo, while cluster II identifies the only element Cd. The cluster pattern may characterise elemental sources as suggested by PCA results. In soil, As and Cd may

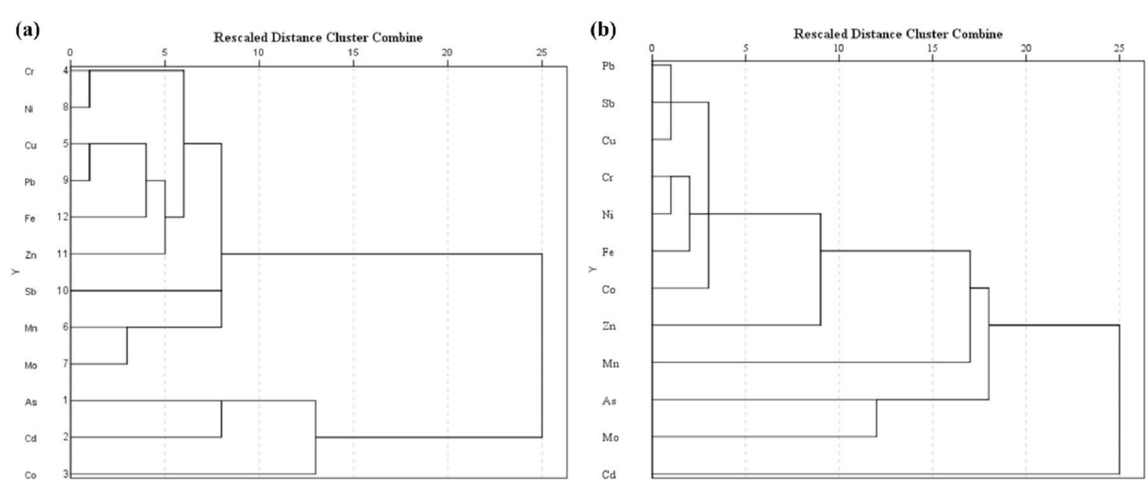

Fig. 3. Dendrogram of cluster analysis (a) soil heavy metals and metalloids, (b) plant foliage heavy metals and metalloids.

have similar origin, however, their diversity of bioavailability in soil may contribute to their difference uptake and translocation in plants (Zhao and Wang, 2020). In addition, plants vary in their sensitivity, resistance, and metabolism to As and Cd (DalCorso et al., 2008; Meharg and Hartley-Whitaker, 2002).

3.2.3. Assessment of potential plant species for phytoremediation

BCF has been used in the present study to measure heavy metal and metalloid accumulation efficiency in plants. The BCF values exceed 1 indicating of potential hyperaccumulator plant species for phytoremediation. As shown in the results in Table 1, Co, Cr and Ni concentrations in soil for all collected sites were under target values according to the Dutch Standard, Fe and Mn which are not included in the Dutch Standard, and they are two necessary nutrients for plants (White and Brown, 2010). Therefore, the relationship of BCF of Co, Cr, Ni, Fe and Mo and their concentrations in soil and plant foliage were not analyzed.

Table 4 showed the screened plant species for phytoremediation from field surveys in the present work and from the published literature. Although there are sites with As, Pb, Sb concentration in soil greater than target values, there is no corresponding plant species with BCF more than 1 were observed. From the field survey, we were able to propose some plant species for As, Pb and Sb remediation based on the BCF values. In addition, for other heavy metals that have concentrations in soil greater than target values, plant species were screened using BCF values of plants greater than 1 as the criteria to select candidates for phytoremediation of Cd, Cu, Mo, Zn. Fig. 4 showed 3D graphs that correlate the relationship between BCF values, concentrations of soil (Cs) and plant foliage (Cp) heavy metals of Cd, Cu, Mo, Zn.

In total, twelve plant species were screened as potential plants for phytoremediation. This group contains one aquatic plant (*Centella asiatica*) growing in high-Cd conditions and three ferns, such as *Pteris vittate* and *Nephrolepis biserrata* with high BCF values for As accumulation and *Dicranopteris linearis* growing in high level Pb soils. Six herbaceous plant species from Asteraceae, Acanthaceae, Moraceae, Fabaceae and Cucurbitaceae familes were selected. The tree *Syzygium grande* was found to grow in high Cu soils in rainforest, which has potential uptake ability for Cu, whereas *Axonopus compressus* (grass species), growing in soils with high concentrations of Cd, Mo and Sb.

The data on the accumulation of the various heavy metal in various plant species found in our work and those that were published was summarized in Table 4. Listed plant species in the present work include well-studied hyperaccumulators. For example, it has been shown that *Pteris vittata* (brake fern) is a hyperaccumulator for As and has been applied in phytoremediation (Ma et al., 2001). It was reported that *Pteris vittata* accumulated the highest As content reached to (19,300 ± 190) mg/kg in a tropical greenhouse after grown for 78 days in contaminated soils (Yong et al., 2010). *Centella asiatica* is one of the Cd hyperaccumulators found in the siding lead-zinc mining area in Liuzhou (a sub-tropical city in China), Guangxi Province (Liu et al., 2016). The accumulation data of some of the tropical plant species were found for the first time and presented in this work. For instance, *Syzygium grande* is good for removal of Cu, *Hemigraphis reptans*, *Desmodium* sp. and *Mukia maderaspatana* are good for Mo, *Fatoua pilosa* is good for Pb, Sb and Zn, *Axonopus compressus* is good for Sb phytoremediation. It has been reported that *Axonopus compressus* as a potential accumulative bio-monitor for Mo in tropical conditions (Tow et al., 2018). *Axonopus compressus* grown in Cd–Zn contaminated soils in Thailand also showed accumulation of Cd in both shoots and roots (Sao et al., 2007), suggesting *Axonopus compressus* could be a multi-elements accumulator for environmental remediation.

4. Conclusion

In this work, twelve heavy metal and metalloid concentrations in soils, that were collected from nature parks and industrial zones in Singapore, were measured and their levels in plant foliage were also investigated. Using the BCF values, the potential of the tropical plant species for phytoremediation were screened which include aquatic plant, ferns, herbaceous plants, tree and grass species. ICP-OES results suggested that As, Cd, Mo, Pb, Zn, Sb, Cu concentrations in soil for some sampling sites were higher than the target values, Co, Cr, Ni levels were lower than the target values in soil from all areas. Cu and Sb in soil in certain site were higher than intervention values based on the Dutch Standard, implying the accumulation of these elements in soils. The results indicated that industrial activities in urban environment have considerable effects on the accumulation and distribution of heavy metals and metalloids in soils, indicating that preventive actions like phytoremediation could be employed to minimize heavy metal contamination in soils on industrial sites. Based on the BCF values of sampling plant foliage for the various heavy metal and metalloids, the accumulation capabilities of twelve tropical plant species, which includes some well-studied one, were screened for potential phytoremediation application. The proposed species are promising candidates for phytoremediation in Singapore.

Table 4
Potential plant species for phytoremediation.

Heavy metal and metalloid	Plant species	Family	[C]s (mg/kg)	[C]p (mg/kg)	BCF	Uptake concentration in plant foliage from literatures (mg/kg)
As	*Pteris vittata*	Pteridaceae	14.37	518.40	36.08	8331.00 (Kalve et al., 2011)
	Nephrolepis biserrata	Nephrolepidaceae	14.37	172.80	12.03	153.96 (Ancheta et al., 2020)
Cd	*Tridax procumbens*	Asteraceae	1.03	1.22	1.18	3.96 (Kumar et al., 2013)
	Centella asiatica	Apiaceae	1.04	1.40	1.35	330.70 (Liu et al., 2016)
	Axonopus compressus	Poaceae	1.02	25.05	24.56	669.00 (Sao et al., 2007)
	Asystasia gangetica	Acanthaceae	1.37	23.68	17.28	0.94 (Kong and Chew, 2014)
Cu	*Fatoua pilosa*	Moraceae	78.68	94.01	1.19	180.00 (Brooks et al., 1978)
	Syzygium grande	Myrtaceae	37.24	41.62	1.12	This study
Mo	*Axonopus compressus*	Poaceae	2.66	6.68	2.51	6000.00 (Tow et al., 2018)
	Hemigraphis reptans	Acanthaceae	7.22	11.36	1.57	This study
	Desmodium sp.	Fabaceae	6.46	69.24	10.72	This study
	Mukia maderaspatana	Cucurbitaceae	4.05	6.19	1.53	This study
Pb	*Dicranopteris linearis*	Gleicheniaceae	29.11	69.85	2.40	60.90 (Chao and Chuang, 2011)
	Fatoua pilosa	Moraceae	36.90	51.97	1.41	This study
	Asystasia gangetica	Acanthaceae	33.82	38.58	1.14	21.79 (Kong and Chew, 2014)
Sb	*Fatoua pilosa*	Moraceae	2.70	2.95	1.09	This study
	Axonopus compressus	Poaceae	0.50	1.57	3.14	This study
Zn	*Fatoua pilosa*	Moraceae	420.92	973.02	2.31	This study
	Asystasia gangetica	Acanthaceae	142.40	468.33	3.29	159.95 (Kong and Chew, 2014)

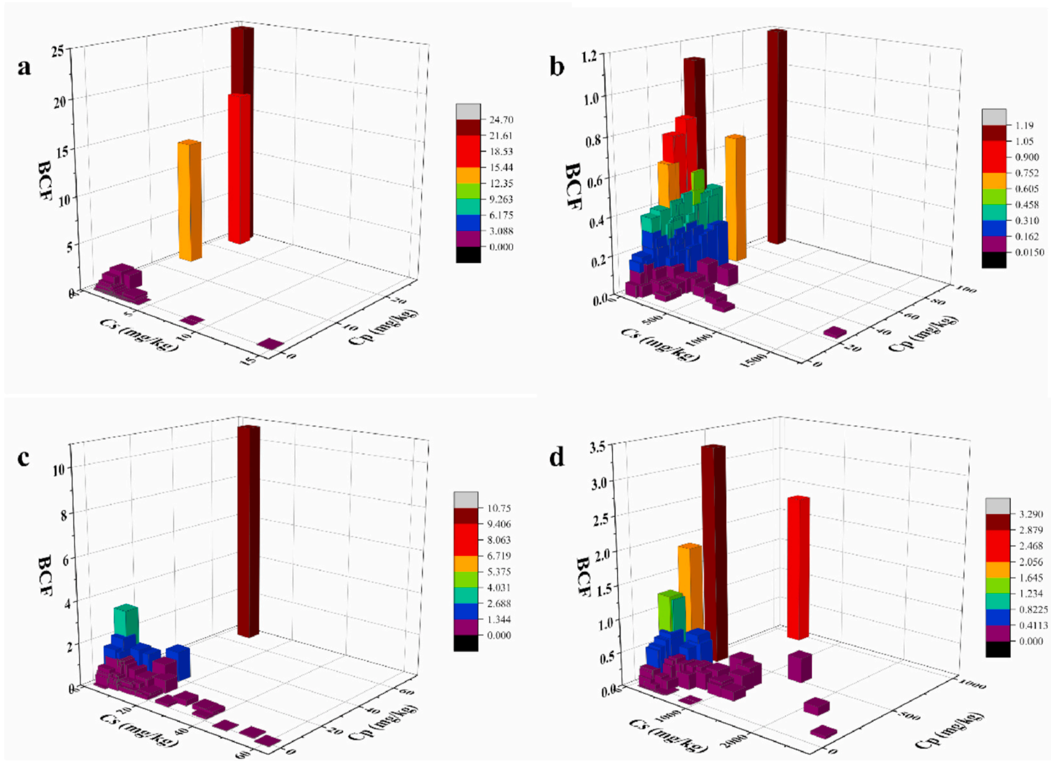

Fig. 4. 3D graph of parameters relationships among Cs, Cp and BCF of (a) Cd, (b) Cu, (c) Mo, (d) Zn, (Cs: soil heavy metal concentrations, Cp: plant foliage heavy metal concentrations).

Authorship contributions

Yamin Wang: Field investigation, Sample collection, preparation and analysis, Writing the original draft, **Swee Ngin Tan**: Plan and design the project, Review and editing the manuscript, **Mohamed Lokman Mohd Yusof**: Field investigation, Plan and design the project, Review and editing the manuscript, Plant species identification, **Subhadip Ghosh**: Field investigation, Plan and design the project, Review and editing the manuscript, Plant species identification, **Yeng Ming Lam**: Plan and design the project, Review and editing the manuscript, Funding acquisition.

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Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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Appendix A. Supplementary data

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