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Soil Factors Influencing Heavy Metal Concentrations in Medicinal Plants

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ABSTRACT

This study was conducted with the aim of finding soil factors which influence heavy metals uptake by medicinal plants. The heavy metal concentrations in medicinal plants at 3 different sites (different soil types) and the soils on which the plants grow were analysed. From the correlation analysis, soil properties affect all of the heavy metal concentrations in soils, meanwhile, only Cu and Se concentrations in soils affect their uptake by plants. However, this depends on plant parts (root and foliar), and the soil types. Principal component analysis (PCA) was also conducted to ascertain any patterns in the soil samples in relation to soil chemical characteristics and reinforce the findings from the correlation analysis. From the principal component analysis in this study, total Pb and As concentrations in medicinal plants were correlated with their concentrations in soils; however, they vary according to the soil types.

Keywords: Heavy metals, medicinal plants, soil properties, agriculture input, correlation analysis, principal component analysis

INTRODUCTION

Medicinal plants play an important and vital role in traditional medicines and are widely consumed as home remedies (Ajasa *et al*., 2004). A survey carried out by World Health Organization (WHO) indicated that about 70-80% of the world population rely on non-conventional medicines, mainly of herbal sources in their primary healthcare (WHO, 2002). The Secretariats of the Convention on Biological Diversity (CBD) also reported US$60 billion sales in the global herbal medicine markets in 2000 (Ang & Lee, 2006). Meanwhile, in Malaysia, the latest

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report shows that the total sales for local herbal products reached approximately RM100 million for the year 2000. In 2005, these sales increased to RM500 million and RM2.5 billion during 2010 (Hassan, 2008). In recent decades, the use of phytopharmaceuticals and herbal medicines has increased worldwide due to several reasons, among them are the side-effects which are often lower than those presented when synthetic drugs are employed, as well as due to the higher costs of many conventional pharmaceutical formulations (Mamani *et al*., 2005). However, the vast majorities of the medicinal herbal products are unlicensed and are not required to demonstrate efficiency, safety or quality. Unknown effects of some of medicinal herbs have been observed. Several examples are allergic reactions, toxic reactions, mutagenic effects, drug interaction, drug contamination, and mistaken plant identities (Basgel & Erdemoglu, 2006).

One obvious safety issue related to the medicinal plants is the possibility that some herbal medicines contain heavy metals (Ernst, 2002). Although the phase three registration of traditional medicines was implemented on 1st January, 1992, there are still many unregistered traditional medicines rampantly available in the Malaysian market with contaminations of heavy metals above the permissible limits. All products registered with the Drug Control Authority (DCA) Malaysia will have to carry a unique product registration number. The unregistered traditional medicines or counterfeits are a serious threat to public health since these drugs are manufactured illegally and contain levels of chemical compounds which, once consumed, could result in serious unknown side effects or even death (Ang, 2008).

Metals are probably the oldest known toxins to man (Mamani *et al*., 2005). The eight most common heavy metal pollutants listed by the Environment Protection Agency (EPA) are arsenic, cadmium, chromium, copper, mercury, nickel, lead and zinc (Athar & Vohora, 1995). Poisonings associated with the presence of toxic metals in medicinal plants have been reported in Asia, Europe and the United States (Olujohungbe *et al*., 1994; Dunbabin *et al*., 1992; Kakosy *et al*., 1996; Markowitz *et al*., 1994). Their basic source, for man, is the food chain (Lozak *et al*., 2002). Ingestion of heavy metals through medicines and food can cause accumulation in organisms, producing serious health hazards such as injury to the kidneys, symptoms of chronic toxicity, renal failure and liver damage (Abou-Arab *et al*., 1999). Individuals generally use herbal medicines for prolonged period to achieve a desirable effect. Prolonged consumption of such herbal medicines may induce chronic or subtle health hazards (Shailendra & Sahadeb, 2002).

Heavy metals in soils originate either from weathering of parent materials and/or from numerous external contaminating sources (Fergusson & Kim, 1991). Plants are an important link for transferring trace elements from soils to man. The level of essential elements in plants varies, in which the content is being affected by the geochemical characteristics of a soil and the ability of plants to selectively accumulate some of these elements. Bioavailability of the elements depends on the nature of their association with the constituents of a soil (Lozak *et al*., 2002). Their availability for plant absorption can be affected by factors including pH, Eh, CEC and organic matter content of soils and concentrations of the competing trace elements (Weiping *et al*., 2008). Plants readily assimilate through the roots such elements which dissolve in water and occur in ionic forms (Lozak *et al*., 2002). High levels of toxic metals can also occur during medicinal preparations or processing when they are used as active ingredients, as in the case of Pb and Hg in some Chinese, Mexican and Indian medicines (Levitt, 1984; Chan *et al*., 1993) or when the plants are grown in polluted areas, such as near roadways or metal mining and smelting operations (Pip, 1991). In addition, high levels can be found when agricultural expedients are used, including cadmium containing fertilizers, organic mercury or lead based pesticides, and contaminated irrigation water (Abou-Arab *et al*., 1999).

In this study, the heavy metals and their concentrations at 3 different sites (different soil types) and the soils on which the plants grow were analysed. The aim of this study was to identify the soil factors affecting heavy metals uptake by correlating the soil factors with the plant data.

MATERIALS AND METHODS

Study Area

The soil and plant samples in this study were randomly collected from the FRIM External Research Station, Setiu, Terengganu (17 samples), Felda Agricultural Services Sdn. Bhd, Jengka 25 Bandar Jerantut, Pahang (16 samples) and MARDI Jalan Kebun, Klang, Selangor (24 samples), with each site having its own management practice. The samples were collected using paired sampling with the soil samples taken adjacent to the medicinal plants which were sampled. At each point, three soil auger borings were composited into one sample. It is noted that each location has a different soil type. For the location at Setiu, Terengganu, the soil type is of the Beach Ridges Interspersed with Swales (BRIS) soil from the Jambu series (Spodic Quartzipsamment), whereas for Jengka 25, Pahang, the soil type is of the Durian series (Plinthaquic Paleudult). As for the location at Jalan Kebun, Klang, the soil is of peaty type.

The history of fertilization for each site also differs. The *Tongkat Ali* collected from Setiu, Terengganu was fertilized with the NPK fertilizer. However, there were also medicinal plants that were unfertilized or considered as growing naturally or wild. The medicinal plants collected from Jengka 25, Pahang were all fertilized with ammonium sulphate (AS), Christmas Island Phosphate Rock (CIRP), muriate of potash (MOP), kieserite and compost. Meanwhile, the medicinal plants from Jalan Kebun, Klang, were fertilized with chicken dung only. The medicinal plants collected were grouped based on the plant parts utilized for medicinal purposes, which are the foliar and roots. This was done in this study because the distribution of heavy metals is different for different plant parts.

Soil and Plant Samples

Aqua-regia digestion (Black *et al*., 1965) was used to extract the total heavy metals in the soil samples, and these elements were determined using the PE 5100 atomic absorption spectrophotometer (AAS). Meanwhile, to determine the total heavy metals in the plant tissue, the dry ashing method (Leo & James, 1973) was used and determined by the Zeeman 4100ZL graphite furnace atomic absorption spectrophotometer. Soil properties data such as pH, EC, CEC, organic carbon (determined by the Walkley and Black method) and clay content (determined by the pipette method) were also required for the correlation analysis, and all the methods were referred to Black *et al*. (1965).

RESULTS AND DISCUSSION

Chemical properties and heavy metal concentrations of the soils under study are given in Table 1 and Table 2, respectively. All of the soils in these 3 locations differed in their total heavy metal concentration.

The soil pH values were below 7.0 (4.52-6.71), indicating the acidic nature of the soils for all the 3 locations. As mentioned by Bang (2002), acidity is the main factor that controls metal mobility in soils. Generally, soils of Malaysia are acidic because they are highly weathered. The total heavy metal concentrations in soils sampled from Jalan Kebun, Klang, which used only the chicken dung fertilizer, were highest for Cu (49.29 mg kg^{-1}), Zn (84.45 mg kg^{-1}), Ni (13.88 mg kg^{-1}), As (12.56 mg kg^{-1}) and Cd (0.72 mg kg^{-1}). Meanwhile, for Jengka 25, Pahang, which used a compound fertilizer for fertilization, had the highest Pb (56.77 mg kg^{-1}) and Se (7.98 mg kg^{-1}) total concentrations. The soil samples collected from Jalan Kebun, Klang, also showed that their chemical properties such as pH, EC, CEC and organic carbon were the highest among all the locations. The sandy BRIS soil sampled from Setiu, Terengganu, exhibited lower concentrations of heavy metals as compared to the other 2 locations.

Tables 3 (a) – (d) show the heavy metal concentrations (mg kg^{-1} dry weight basis) in the medicinal plants sampled from Setiu, Terengganu, Jengka 25, Pahang and Jalan Kebun, Klang. The concentrations obtained were compared to the maximum permissible concentrations as stated in the Malaysian Herbal Monograph (2009), which are 10, 5.0 and 0.3 mg kg^{-1} for Pb, As and Cd, respectively. On the other hand, for Cu and Zn, the concentration values were compared to the maximum permissible concentrations as stated in the Malaysian Food Act (1983) and Malaysian Food Regulation (1985).

Based on the data as shown in Table 3(a), in comparing between the *Tongkat Ali* from Setiu, Terengganu and Jengka 25, Pahang, the *Tongkat Ali* from the latter site tended to have higher heavy metal concentrations except for Ni and Se. However, in terms of maximum permissible concentrations,

TABLE 1
Chemical properties of the soils at different locations.

Locations	Soil properties				
	pH	EC ($dScm^{-1}$)	CEC ($cmolkg^{-1}$)	Organic Carbon(%)	Clay Content (%)
Setiu, Terengganu	3.49 – 5.22	10.4 – 88.8	1.26 – 13.0	0.46 – 6.70	0 – 1.64
Mean ± SE, n=17	4.37 ± 0.13	30.87 ± 5.92	5.39 ± 0.92	3.37± 0.58	0.27 ± 0.12
Jengka 25, Pahang	4.22 – 6.43	23.4 – 217.3	2.45 – 15.06	0.05 – 2.09	21.04 – 57.01
Mean ± SE, n=16	5.42 ± 0.19	74.42 ± 13.05	7.71 ± 0.98	0.73 ± 0.11	33.30 ± 3.13
Jalan Kebun, Klang, Selangor	5.98 – 7.30	235 - 2780	21.40 – 59.10	4.74- 6.38	0
Mean ± SE, n=24	6.71 ± 0.13	654.97 ± 135.28	42.31 ± 2.04	5.91 ± 0.06	0

SE: Standard Error

TABLE 2
Total heavy metals concentration ($mgkg^{-1}$) in the soil samples at different locations.

Locations	Heavy metals concentration in soil ($mgkg^{-1}$)*						
	Cu	Zn	Ni	Pb	As	Cd	Se
Setiu, Terengganu	1.87 – 3.73	5.73 – 11.47	0.28 – 4.33	0.70 – 4.35	0.16 – 2.86	0.01 – 0.42	0.06 – 0.78
Mean ± SE, n=17	2.60 ± 0.13	7.80 ± 0.44	1.63 ± 0.32	2.88 ± 0.42	1.09 ± 0.22	0.09 ± 0.02	0.28 ± 0.05
Jengka 25, Pahang	4.53 – 21.20	12.67 – 59.07	3.65 – 9.26	29.01 – 118.52	0 – 26.27	0.11 – 0.85	0.39 – 51.83
Mean ± SE, n=16	12.07 ± 1.23	28.19 ± 3.70	6.37 ± 0.36	56.77 ± 6.91	9.72 ± 2.95	0.46 ± 0.07	7.98 ± 4.18
Jalan Kebun, Klang, Selangor	33.04 – 69.72	50.76 – 141.12	7.32 – 17.87	1.50 – 25.36	4.37 – 36.65	0.36 – 0.93	0.18 – 2.29
Mean ± SE, n=24	49.29 ± 2.06	84.45 ± 4.67	13.88 ± 0.52	7.15 ± 1.20	12.56 ± 1.56	0.72 ± 0.03	1.07 ± 0.11

SE: Standard Error
*95th percentile concentration levels for:
Cu: 50 $mgkg^{-1}$; Zn: 95 $mgkg^{-1}$; Ni: 45 $mgkg^{-1}$; Pb: 65 $mgkg^{-1}$; As: 60 $mgkg^{-1}$; and Cd: 0.30 $mgkg^{-1}$

Cd concentration in a few of the *Tongkat Ali* samples from Setiu, Terengganu and all of the *Tongkat Ali* samples from Jengka 25, Pahang exceeded the reference limit. Other than that, Pb concentration in most of the *Tongkat Ali* sampled from Jengka 25, Pahang, was also found to exceed the reference limit.

Table 3(b) shows the heavy metal concentrations in the leafy medicinal plants sampled from Setiu, Terengganu. As observed, Cu, Zn, Pb and Cd concentrations in certain medicinal plants already exceeded the maximum permissible concentrations. For the heavy metal concentrations in the leafy medicinal plants sampled from Jengka

TABLE 3(a)
Heavy metal concentrations (mgkg-1, dry weight basis) in Tongkat Ali (*Eurycoma longifolia*) samples at different locations.

	Sample	Elements*						
		Cu	Zn	Ni	Pb	As	Cd	Se
Setiu, Terengganu	Tongkat Ali Control R2	9.33	22.33	0.00	2.93	0.05	0.00	2.29
	Tongkat Ali Control R3	10.33	13.00	1.92	2.08	0.10	0.00	2.68
	Tongkat Ali Control R4	10.67	22.33	0.98	1.20	0.10	0.00	2.39
	Tongkat Ali NPK R1	9.33	24.33	21.85	1.27	0.01	0.00	1.75
	Tongkat Ali NPK R2	8.00	22.00	10.91	1.68	0.23	0.52	0.22
	Tongkat Ali NPK R3	8.33	26.33	10.41	1.97	0.22	2.06	0.42
	Tongkat Ali NPK R4	9.67	25.67	0.00	1.29	0.10	2.88	0.06
	Tongkat Ali (W)	9.67	15.00	0.00	3.97	0.05	0.00	0.48
	Mean ± SE	9.42 ± 0.32	21.37 ± 1.72	5.76 ± 2.82	2.06 ± 0.34	0.11 ± 0.03	0.68 ± 0.40	1.2 ± 0.39
Jengka 25, Pahang	Tongkat Ali R1	16.00	23.50	3.07	22.07	0.70	1.11	0.02
	Tongkat Ali R2	15.00	34.00	2.83	4.73	0.32	0.77	0.18
	Tongkat Ali R3	17.00	35.00	2.07	50.15	0.34	1.38	0.16
	Tongkat Ali R4	13.00	26.50	3.54	11.07	0.33	0.68	0.16
	Mean ± SE	15.25 ± 0.85	29.75 ± 2.82	2.88 ± 0.31	22.0 ± 0.04	0.42 ± 0.09	0.99 ± 0.16	0.13 ± 0.04

SE: Standard Error
*Maximum permitted concentration for:
Cu: 30 mgkg-1; Zn: 40 mgkg-1; Pb: 10 mgkg-1; As: 5.0mgkg-1 and Cd: 0.3 mgkg-1.

TABLE 3(b)
Heavy metal concentrations (mgkg-1, dry weight basis) in the leafy medicinal plants sampled from Setiu, Terengganu

Sample	Elements						
	Cu	Zn	Ni	Pb	As	Cd	Se
Cucur Atap (*Baeckea frutescens*) (W)	25.53	33.00	6.91	12.43	0.28	0.48	2.78
Kerbau Amok (*Schefflera ridleyi*) (W)	27.63	30.67	3.22	5.89	0.43	1.03	3.28
Mata Ayam (*Ardisia crenata*) (W)	25.93	44.00	2.99	11.62	1.51	0.22	1.35
Mas Cotek (*Ficus deltoidea*) (W)	21.53	174.7	4.05	15.70	0.45	0.20	0.60
Senduduk (*Melastoma malabathricum*) 1 (W)	41.23	46.33	3.23	35.09	1.70	0.43	0.65
Senduduk (*Melastoma malabathricum*) 2 (W)	39.17	102.7	2.47	7.88	1.14	0.34	0.87
Gelam (*Melaleuca cajputi*) (W)	21.37	35.00	3.15	8.89	0.87	0.32	1.92

TABLE 3(b) (continue)

Kapal terbang (*Chromolaena odorata*) (W)	75.90	60.00	5.45	13.41	1.66	2.18	0.88
Kemunting (*Catharanthus roseus*) (W)	21.40	63.33	2.67	8.38	0.29	0.21	5.20
Mean ± SE	33.30 ± 5.87	65.52 ± 15.53	3.79 ± 0.49	13.25 ± 2.92	0.93 ± 0.20	0.60 ± 0.21	1.95 ± 0.52

SE: Standard Error (W): Wild plant
*Maximum permitted concentration for:
Cu: 30 $mgkg^{-1}$; Zn: 40 $mgkg^{-1}$; Pb: 10 $mgkg^{-1}$; As: 5.0 $mgkg^{-1}$; and Cd: 0.3 $mgkg^{-1}$

TABLE 3(c)
Heavy metal concentrations ($mgkg^{-1}$, dry weight basis) in the leafy medicinal plants from Jengka 25, Pahang.

Sample	Elements						
	Cu	Zn	Ni	Pb	As	Cd	Se
Kacip Fatimah (*Labisia Pumila*) R1	18.00	38.50	1.43	6.58	0.59	1.47	0.44
Kacip Fatimah R2	14.50	27.00	2.64	2.01	1.33	1.80	0.60
Kacip Fatimah R3	15.00	26.00	2.24	5.82	0.30	1.94	0.20
Kacip Fatimah R4	16.00	24.50	2.76	2.58	0.18	0.19	0.25
Mas Cotek (*Ficus deltoidea*) R1	16.00	16.50	0.96	1.46	0.33	1.37	0.15
Mas Cotek R2	12.50	14.50	1.01	2.39	0.25	0.12	0.23
Mas Cotek R3	9.50	12.00	0.66	2.08	0.28	0.08	0.03
Mas Cotek R4	9.50	15.00	0.45	2.48	0.45	0.09	0.19
Misai Kucing (*Orthasiphon stamineus*) R1	17.50	35.00	0.68	2.79	0.00	0.09	0.01
Misai Kucing R2	18.00	38.50	1.67	0.55	0.00	0.09	0.11
Misai Kucing R3	24.00	42.50	1.96	3.02	0.00	0.10	0.13
Misai Kucing R4	19.00	32.00	5.71	4.79	0.00	0.14	0.37
Mean ± SE	15.79 ± 1.18	26.83 ± 3.06	1.85 ± 0.42	3.05 ± 0.52	0.31 ± 0.11	0.62 ± 0.22	0.23 ± 0.05

SE: Standard Error
*Maximum permitted concentration for:
Cu: 30 $mgkg^{-1}$; Zn: 40 $mgkg^{-1}$; Pb: 10 $mgkg^{-1}$; As: 5.0 $mgkg^{-1}$; and Cd: 0.3 $mgkg^{-1}$.

TABLE 3(d)
Heavy metal concentrations ($mgkg^{-1}$, dry weight basis) in the medicinal plants from Jalan Kebun, Klang, Selangor.

Sample	Elements						
	Cu	Zn	Ni	Pb	As	Cd	Se
Ginseng Jepun (*Panax japonica*)	8.35	56.70	0.10	0.74	0.96	0.14	0.79
Tetulang @ hempedu ular	7.95	35.80	0.00	0.61	0.88	0.06	0.46
Ketumbar jawa (*Eryngium foetidum L.*)	10.35	40.15	1.65	0.82	0.58	0.08	0.04

TABLE 3(d) (continue)

Naga buana (*Phyllanthus pulcher*)	11.10	48.30	4.24	1.29	0.37	0.08	0.31
Tebu badak	8.60	29.20	2.88	1.71	0.36	0.13	0.05
Kunyit hantu (*Curcuma aeruginosa*)	5.45	17.70	3.08	1.27	0.86	0.09	0.11
Asparagus (*Asparagus officinalis*)	6.80	37.85	2.80	0.14	0.49	0.01	0.00
Lemayung hijau (*Zingiber zerumbet*)	6.10	40.85	2.29	0.28	0.22	0.18	0.44
Pegaga brunei (*Centella asiatica*)	5.25	34.90	2.50	0.68	0.53	0.08	0.15
Daun Mengkudu (*Morinda citrifolia*)	6.35	33.90	2.58	0.93	0.20	0.10	0.19
Kaduk (*Piper sarmentosum roxb*)	10.15	42.10	2.73	0.82	0.21	0.09	0.87
Bangun-bangun (*Solenostemon amboinicus*)	8.10	56.70	2.31	1.28	0.38	0.07	0.45
Jintan hitam (*Nigella Sativa*)	2.70	27.20	2.51	0.55	0.64	0.36	0.23
Sabung nyawa (*Gynura procumbens*)	7.15	31.55	2.83	0.26	0.51	0.09	0.11
Tembaga suasa (*Hanguana malayana (Jack) Merr*.)	5.70	49.50	0.81	0.59	0.33	0.12	0.00
Cekur jantan (*Kaempferia galangal*)	3.15	27.95	0.71	0.77	1.27	0.12	0.00
Selasih serai (*Ocimum Sanctum*)	4.05	87.55	0.89	1.03	0.70	0.25	0.00
Pegaga melayu (*Centella asiatica*)	4.10	79.10	0.73	0.59	0.41	0.70	0.00
Pegaga nyonya (*Centella asiatica*)	3.75	29.55	0.92	5.52	0.70	0.54	0.00
Cekur mas (*Kaempferia galangal*)	8.80	25.85	0.83	5.18	0.42	0.26	0.00
Mata pelanduk (*Ardisia crenata*)	6.65	31.80	0.88	0.30	1.06	0.08	0.00
Pudina (*Mentha arvensis*)	14.10	34.90	0.91	0.54	0.86	0.14	0.00
Beremi (*Limnophila aromatic*)	9.00	37.95	0.97	0.34	0.49	0.08	0.27
Kesum (*Polygonum minus huds*)	6.90	22.85	4.18	4.05	1.27	0.13	0.00
Mean ± SE	7.11 ± 0.56	40 ± 3.38	1.85 ± 0.25	1.26 ± 0.30	0.61 ± 0.06	0.17 ± 0.03	0.19± 0.05

SE: Standard Error
*Maximum permitted concentration for:
Cu: 30 $mgkg^{-1}$; Zn: 40 $mgkg^{-1}$; Pb: 10 $mgkg^{-1}$; As: 5.0 $mgkg^{-1}$; and Cd: 0.3 $mgkg^{-1}$.

25, Pahang, Table 3(c) shows that only a few medicinal plants exceeded the limit for Cd concentration. Conversely for the other samples, their heavy metal concentrations were still under the permissible levels. Table 3(d) shows the heavy metal concentrations in the leafy medicinal plants from Jalan Kebun, Klang. From the table, it shows that some of the medicinal plants exceeded the maximum permissible concentrations, which were Zn and Cd.

Correlation analysis was carried out in order to see the relationship between heavy metal concentrations in the soil samples with the soil properties, or the soil factors that influence or control heavy metal concentrations in soils. The correlation analysis between the soil properties (i.e.

pH, EC, CEC, organic carbon, and clay content) with heavy metal concentrations in soils grown with the *Tongkat Ali* are shown in Table 4 (a). As observed in this table, Cu and Zn concentrations in the soils sampled from Jengka 25, Pahang were negatively correlated with CEC and organic carbon of the soils, respectively. Notably, arsenic concentration exhibited a positive correlation with organic carbon of the soils, while the other elements were not significantly correlated with any soil properties of the soils from Jengka 25, Pahang. Table 4 (b) shows the correlation coefficients between soil properties with heavy metal concentrations in soils grown with leafy medicinal plants. From this table, the pH values of soils from Setiu, Terengganu were found to affect most of heavy metal concentrations (Cu, Ni, Pb, As and Se) in the soil samples from there. Furthermore, organic carbon was also found to affect Pb concentration inversely. For the soil samples from Jengka 25, Pahang, all the soil properties were found to affect heavy metal concentrations in the soil samples. As observed, the pH of the soils affected Cu, Zn, Pb and Cd concentrations positively. Moreover, electrical conductivity also affected Zn, Ni, Pb and Cd concentrations of the soils from Jengka 25. Besides, clay content was also positively correlated with Cu, Zn, Ni, Pb and Cd concentrations in the soils from Jengka 25, Pahang, whereas organic carbon showed a positive correlation with Se concentration in that soils. For the site at Jalan Kebun, Klang, the properties of the soils did not show any significant correlation with heavy metal concentrations in the soils, except for EC, which was positively correlated with As concentration in the peaty type soil.

TABLE 4(a)
Correlation coefficients (r) between soil properties with heavy metals concentration in soil samples grown with Tongkat Ali (*Eurycoma longifolia*) at different locations.

Location	Soil properties	Correlation coefficients, r						
		Cu	Zn	Ni	Pb	As	Cd	Se
Setiu, Terengganu (n=8)	pH	0.01	-0.54	0.15	0.07	0.44	-0.16	0.41
	EC	0.20	0.26	-0.09	-0.12	-0.34	0.17	-0.03
	CEC	0.30	0.41	-0.14	-0.17	-0.42	0.11	-0.01
	Organic carbon	0.30	0.43	-0.24	-0.25	-0.52	0.08	-0.02
Jengka 25, Pahang (n=4)	pH	0.21	0.42	0.44	0.96	-0.37	0.13	-0.75
	EC	-0.74	-0.88	-0.13	-0.59	0.80	-0.32	0.65
	CEC	-0.99*	-0.93	0.10	0.16	0.77	-0.59	0.01
	Organic carbon	-0.92	-0.98*	0.28	-0.18	0.97*	-0.22	0.51
	Clay content	0.23	0.11	-0.91	-0.78	-0.32	-0.38	0.17

* Significantly correlated ($p<0.05$)

TABLE 4(b)
Correlation coefficients (r) between soil properties with heavy metal concentrations in the soil samples grown with the leafy medicinal plants at different locations.

Location	Soil properties	Correlation coefficients, r						
		Cu	Zn	Ni	Pb	As	Cd	Se
Setiu, Terengganu (n=9)	pH	0.34	0.67*	0.75*	0.85*	0.86*	0.64	0.81*
	EC	-0.33	-0.30	-0.31	-0.51	-0.33	-0.38	-0.39
	CEC	-0.37	-0.50	-0.39	-0.58	-0.54	-0.45	-0.43
	Organic carbon	-0.55	-0.54	-0.19	-0.69*	-0.51	-0.61	-0.59
	Clay	-0.09	0.09	0.01	0.06	0.13	0.06	-0.22
Jengka 25, Pahang (n=12)	pH	0.71*	0.66*	0.53	0.75*	0.38	0.79*	-0.04
	EC	0.50	0.84*	0.72*	0.73*	0.46	0.73*	-0.19
	CEC	0.08	-0.26	-0.55	-0.19	-0.37	0.05	-0.03
	Organic carbon	-0.12	0.03	0.08	0.26	-0.11	0.24	0.84*
	Clay content	0.63*	0.85*	0.73*	0.88*	0.28	0.73*	-0.16
Jalan Kebun, Klang (n=24)	pH	0.01	0.03	-0.09	-0.01	-0.14	0.26	-0.25
	EC	0.14	0.23	-0.02	0.20	0.63*	0.13	0.004
	CEC	0.34	0.13	-0.40	0.08	-0.15	-0.16	-0.37
	Organic Carbon	0.12	0.17	-0.09	-0.09	0.12	0.09	0.05

* Significantly correlated ($p<0.05$)

The correlation analyses between the heavy metal concentrations in the *Tongkat Ali* and leafy medicinal plants with soil properties (pH, EC, CEC, organic carbon, clay content) are shown in Tables 5(a) and(b), respectively. From Table 5(a), the pH value of the soils from Setiu, Terengganu was positively correlated with Zn concentration in the *Tongkat Ali*, which means Zn concentration in the plants would increase with the increase in the soil pH. Meanwhile, Pb concentration in the plants increased with the increase in EC and CEC of the BRIS soil. For the *Tongkat Ali* sampled from Jengka 25, Pahang, the EC of the soils were found to have a positive correlation with As and a negative correlation with Se concentration in the *Tongkat Ali*. The correlation analysis between the soil properties with heavy metal concentrations in the leafy medicinal plants is shown in Table 5(b). From the table, there was no correlation observed between the soil properties with heavy metal concentrations in the leafy medicinal plants collected from Setiu, Terengganu. Meanwhile, for the medicinal plants collected from Jengka 25, Pahang, Pb concentration showed a positive correlation with pH and EC of the soils. Other than EC and pH of the soils, clay content in the soils of Jengka 25, Pahang, also affected the Cd concentration in the

TABLE 5(a)
Correlation coefficients (r) between soil properties with heavy metal concentrations in Tongkat Ali (*Eurycoma longifolia*) at different locations.

Location	Soil properties	Correlation coefficients, r						
		Cu	Zn	Ni	Pb	As	Cd	Se
Setiu, Terengganu (n=8)	pH	-0.49	0.72*	0.32	-0.43	0.36	0.30	0.06
	EC	0.19	-0.57	-0.31	0.76*	-0.29	-0.24	-0.25
	CEC	0.24	-0.66	-0.32	0.72*	-0.23	-0.20	-0.21
	Organic carbon	0.26	-0.59	-0.26	0.63	-0.22	-0.16	-0.23
Jengka 25, Pahang (n=4)	pH	0.40	0.86	-0.79	0.65	-0.70	0.47	0.69
	EC	0.15	-0.83	0.34	-0.08	0.98*	0.14	-0.98*
	CEC	0.82	-0.22	-0.45	0.56	0.79	0.79	-0.78
	Organic carbon	0.45	-0.65	-0.01	0.39	0.92	0.55	-0.94
	Clay content	-0.43	-0.30	-0.59	-0.91	0.21	-0.71	-0.15

* Significantly correlated ($p<0.05$)

TABLE 5(b)
Correlation coefficients (r) between soil properties with heavy metal concentrations in the leafy medicinal plants at different locations.

Location	Soil properties	Correlation coefficients, r						
		Cu	Zn	Ni	Pb	As	Cd	Se
Setiu, Terengganu (n=9)	pH	0.53	-0.30	0.27	0.14	0.41	0.47	-0.33
	EC	-0.23	-0.05	-0.51	-0.04	0.21	-0.23	-0.02
	CEC	-0.34	0.46	-0.40	-0.17	0.01	-0.47	-0.19
	Organic carbon	-0.15	-0.01	0.37	0.46	-0.10	-0.05	-0.04
	Clay	0.07	0.14	-0.49	0.09	0.19	-0.11	-0.33
Jengka 25, Pahang (n=12)	pH	0.15	0.47	0.30	0.63*	0.32	0.40	0.48
	EC	0.09	0.22	0.20	0.58*	0.18	0.55	0.35
	CEC	0.53	0.39	0.45	-0.45	-0.44	-0.43	-0.22
	Organic carbon	-0.14	0.12	-0.44	-0.26	-0.01	-0.07	-0.29
	Clay content	0.09	0.21	0.36	0.44	0.49	0.64*	0.58
Jalan Kebun, Klang (n=24)	pH	0.12	-0.35	0.12	0.13	0.01	-0.33	0.34
	EC	0.016	-0.06	0.04	0.14	-0.17	0.22	-0.26
	CEC	-0.35	-0.33	0.41*	0.38	-0.14	0.05	-0.16
	Organic Carbon	-0.51*	0.09	0.17	0.07	-0.37	0.05	0.06

* Significantly correlated ($p<0.05$)

medicinal plants. The cation exchange capacity of the soils from Jalan Kebun, Klang, also revealed a positive correlation with Ni concentration in the leafy medicinal plants collected from there, while Cu concentration was negatively correlated with organic carbon of the soils.

The correlation analyses between the total heavy metal concentrations in the soils with the heavy metal concentrations in the *Tongkat Ali* and leafy medicinal plants are shown in Tables 6(a) and (b), respectively. In the *Tongkat Ali* samples, as shown in Table 6(a), there was no correlation between the heavy metal concentrations in the soils and in the *Tongkat Ali* being observed. Meanwhile, as revealed in Table 6(b), the data shows that only Cu and Se concentrations in the soils collected from Jalan Kebun, Klang, had a correlation with their concentration in the medicinal plants.

The principal component analysis (PCA) of the soil samples (Fig.1) shows that Cu concentration in the soils was correlated

TABLE 6(a)
Correlation coefficients (r) between heavy metal concentrations in soil with heavy metals concentrations in Tongkat Ali (*Eurycoma longifolia*) at different locations.

Correlation coefficients, r	Location	
	Setiu, Terengganu	Jengka 25, Pahang
Cu in soil with Cu in plant	-0.26 ^{ns}	-0.75 ^{ns}
Zn in soil with Zn in plant	-0.59 ^{ns}	0.57 ^{ns}
Ni in soil with Ni in plant	0.08 ^{ns}	-0.21 ^{ns}
Pb in soil with Pb in plant	0.03 ^{ns}	0.82 ^{ns}
As in soil with As in plant	0.29 ^{ns}	0.83 ^{ns}
Cd in soil with Cd in plant	0.59 ^{ns}	-0.33 ^{ns}
Se in soil with Se in plant	-0.46 ^{ns}	-0.59 ^{ns}

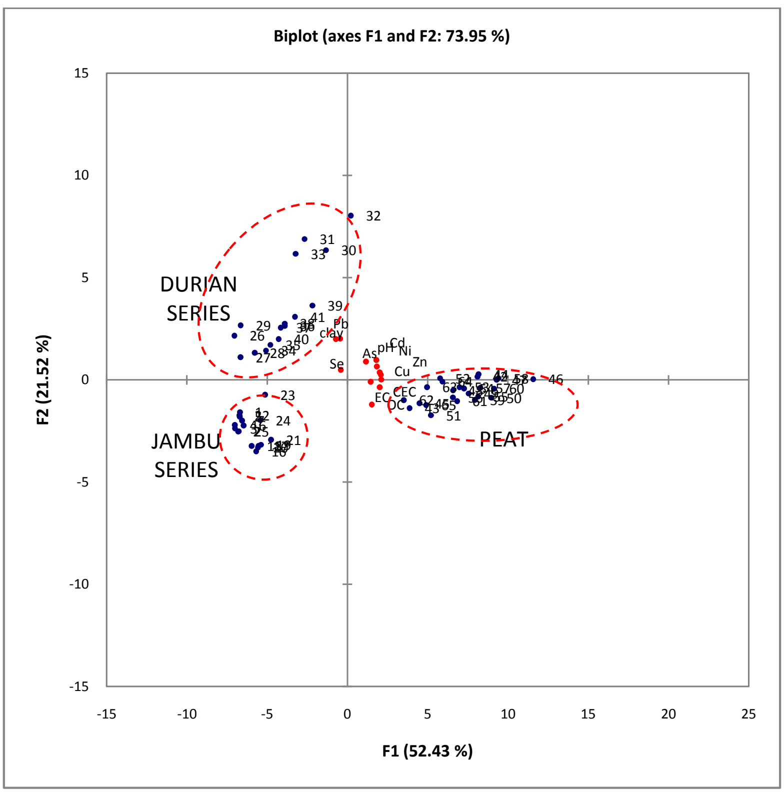
^{ns} Not significant ($p>0.05$)

TABLE 6(b)
Correlation coefficients (r) between heavy metal concentrations in the soil with heavy metal concentrations in the leafy medicinal plants at different locations.

Correlation coefficients, r	Locations		
	Setiu, Terengganu	Jengka 25, Pahang	Jalan Kebun, Klang, Selangor
Cu in soil with Cu in plant	0.60	0.52	-0.45*
Zn in soil with Zn in plant	-0.23	0.13	0.40
Ni in soil with Ni in plant	0.043	0.18	-0.25
Pb in soil with Pb in plant	0.02	0.25	-0.01
As in soil with As in plant	0.66	-0.01	-0.33
Cd in soil with Cd in plant	-0.18	0.29	0.36
Se in soil with Se in plant	-0.31	-0.34	-0.48*

* Significantly correlated ($p<0.05$)

with EC, CEC and organic carbon of the soils, according to the quartile of which it fell into. Whereas, most heavy metals like Zn, Ni, Cd and As were correlated with the pH of the soils. Selenium and Pb in the soils were shown to be correlated with each other, and its concentration was affected by the clay content of the soils. Soil types also had an influence on the heavy metal concentrations in the soil samples. As observed in Fig.1, the soil samples from Jalan Kebun, Klang tended to have high levels of Cu, Zn, Ni, As, and Cd. Besides, the peaty type soils from Jalan Kebun, Klang, also recorded the highest levels of pH, EC, CEC and organic carbon. Meanwhile, the Durian series soil samples from Jengka 25, Pahang, showed the highest levels of clay, Pb and Se, whereas the BRIS soil from Setiu, Terengganu, was characterized to have relatively lower values of heavy metal concentrations and soil properties.



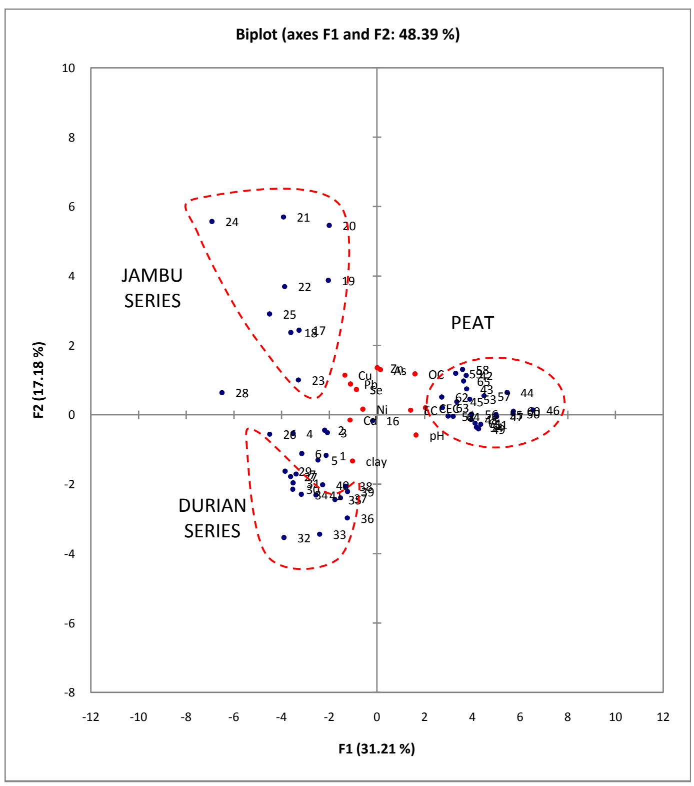
Legend:
pH: Soil pH | EC: Soil EC | CEC: Soil CEC | OC: Organic Carbon
Clay: Clay content | Cu: Cu conc. in soil | Zn: Zn conc. in soil
Ni: Ni conc. in soil | Pb: Pb conc. in soil | As: As conc. in soil
Cd: Cd conc. in soil | Se: Se conc. in soil

SAMPLES FROM:
1-25: Jambu series soil | 26-41: Durian series soil | 42-65: Peat

Fig.1: The principal component plot of the heavy metal concentrations in the soil and chemical properties of the soils sampled in this study

The principal component analysis (PCA) between the soil properties with the heavy metal concentrations in the medicinal plants was also conducted (Fig.2). From this analysis, Zn and As concentrations in the medicinal plants were influenced by pH, EC, CEC and OC of the soils, and this can be clearly observed in the samples from Jalan Kebun, Klang. In addition, cadmium concentration in the medicinal plants sampled from Jengka 25, Pahang grown in the Durian series soil was also found to correlate with the clay content of the soils.

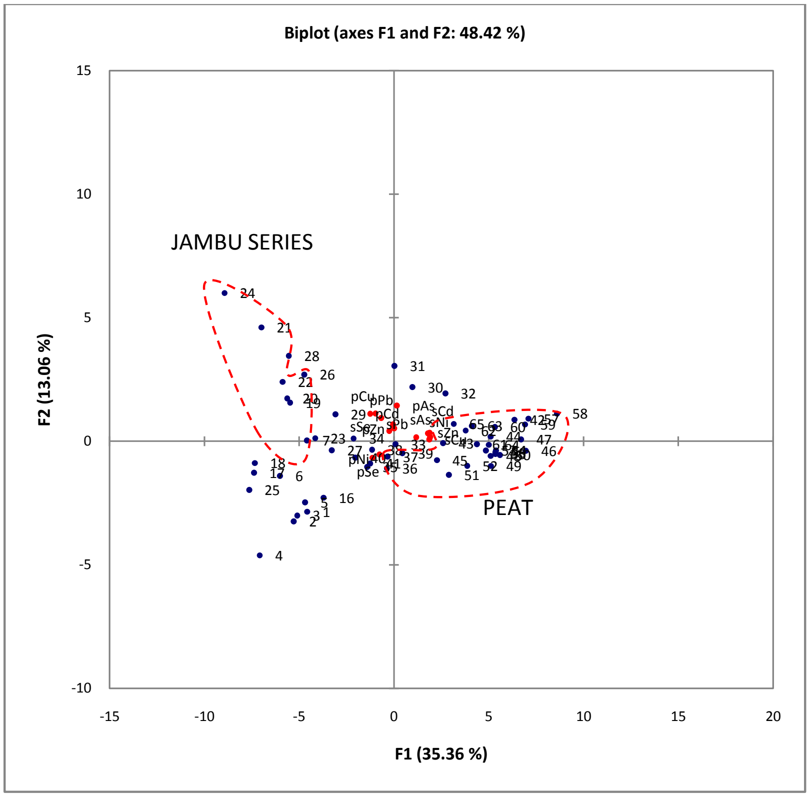
The principal component analysis (PCA) of the heavy metal concentrations in the medicinal plants and soils (Fig.3) showed that only As concentration in the soils was correlated with its concentration in the medicinal plants, and this applies to the medicinal plants grown in the peat soil from Jalan Kebun, Klang. Furthermore, from this analysis, Pb concentration in the



Legend:
pH: Soil pH | EC: Soil EC | CEC: Soil CEC | OC: Organic Carbon
Clay: Clay content | Cu: Cu conc. in plants | Zn: Zn conc. in plants
Ni: Ni conc. in plants | Pb: Pb conc. in plants | As: As conc. in plants
Cd: Cd conc. in plants | Se: Se conc. in plants

SAMPLES FROM:
1-25: Jambu series soil | 26-41: Durian series soil | 42-65: Peat

Fig.2: The Principal component plot of the heavy metal concentrations in the medicinal plant and chemical properties of the soils sampled in this study



Legend:

pCu: Cu conc. in plant	pZn: Zn conc. in plant	pNi: Ni conc. in plant
pPb: Pb conc. in plant	pAs: As conc. in plant	pCd: Cd conc. in plant
pSe: Se conc. in plant	sCu: Cu conc. in soil	sZn: Zn conc. in soil
sNi: Ni conc. in soil	sPb: Pb conc. in soil	sAs: As conc. in soil
sCd: Cd conc. in soil	sSe: Se conc. in soil	

SAMPLES FROM:
1-25: Jambu series soil   26-41: Durian series soil   42-65: Peat

Fig.3: The principal component score plot of the heavy metal concentrations in the medicinal plants and heavy metal concentrations in the soils sampled in this study

soils was also found to correlate with its concentration in the medicinal plants, and this applies to the samples collected from Setiu, Terengganu, and a few plant samples from Jengka 25, Pahang, grown on the Durian series soil.

CONCLUSION

In this study, the random sampling of the medicinal plants was necessary in order to investigate the sources of heavy metals. The sources of medicinal plants nowadays are no more found from natural resources or those that grow in the wild since they have been domesticated. Thus, apart from the soils where the medicinal plants are planted, the sources of heavy metals can also come from fertilizers, pesticides or other sources. In this research, from all the 3 study areas where the medicinal plants were collected, each area had different levels of heavy metal concentrations. All the locations was found to have different soil types, therefore, they varied in soil properties. The principal component analysis between the soil properties with the heavy metal concentrations in the medicinal plants showed that all the soil properties determined affected Zn, As and Cd concentrations in the medicinal plants, which depended on the soil types where the plants were grown. From the principal component analysis in this study, the concentrations of Pb and As in the plants were also correlated with the heavy metals in soils; however, there was a variation according to soil types. Notably, various plants have their own ability to take up heavy metals from soils. Hence, this ability may have contributed to the elevated level of heavy metal concentrations in medicinal plants, other than their concentration in soils.

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