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Relationship between Metals in Vegetables with Soils in Farmlands of Kuching, Sarawak

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ABSTRACT

The accumulation of Fe, Zn, Cu, Mn, Co and Pb in vegetables was investigated in two farmlands, Siburan and Beratok at Kuching, Sarawak. Leafy and fruit vegetable samples were collected and analysed for metal content using the acid wet digestion method. Topsoil samples (0-30 cm) were characterised for pH, organic matter, particle size, nitrogen content, phosphorus content and heavy metals. Leafy vegetables accumulated higher amount of metals compared to fruit vegetables. Metals were determined at the highest concentrations in leaves of kale at Beratok compared to other plant parts. No clear pattern of metal uptake in different parts was observed for green mustard and white mustard. Essential metals, Fe, Zn and Cu, were generally high in the vegetables. Pb levels in the vegetables analysed (dry weight basis) exceeded slightly the level recommended by the Malaysian Food Act 1983. Atmospheric deposition and gas emissions from traffic were the contributing factors for Pb contamination since Pb displayed a tendency to accumulate in leaves compared to other parts of the vegetables. Element concentrations in the soils differed between sampling sites. Correlation analysis yielded a significant relationship between Zn concentrations in soils and vegetables ($n = 15$, $r = 0.86$, $P = 0.001$) and moderate correlation for Cu ($r = 0.55$, $P<0.05$) and Pb ($r = 0.65$, $P<0.05$).

Keywords: Metals, vegetables, soil, correlation

INTRODUCTION

Vegetables are vital to human diet as they contain essential components needed by the human body such as carbohydrates, proteins, vitamins, minerals and also trace elements (Itanna 2002). Consumption of vegetables is one of the pathways by which heavy metals enter the food chain (Wang *et al.* 2004). Excessive accumulation of dietary heavy metals such as Cd, Pb and Cr can lead to serious health problems. Heavy metals persist in the environment, are non-biodegradable and have the potential to accumulate in different body organs (Radwan and Salama 2006). The process of plant growth depends on the cycle of nutrients including trace elements from soil to plant (Mohamed *et al*. 2003).

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Various studies have been conducted on metal assessment in edible vegetables, crops and also soils. These studies have shown the ability of crops to take up heavy metals (Stalikas *et al.* 1997; Mohamed *et al.* 2003; Sharma *et al.* 2007). A study done on edible green vegetables grown along sites of the Sinza and Msimbazi rivers of Tanzania revealed that some of the vegetables contained high levels of metals beyond the levels designated by FAO/WHO (Bahemuka and Mubofu. 1999). Alam *et al.* (2002) showed that vegetables in Samta Village in Bangladesh exhibited the potential to accumulate high levels of Pb that would pose a health hazard for human consumption. A study by Cui *et al.* (2004) reported that vegetables and soils from villages located at 1500 m and 500 m from a smelter in Naning, China were heavily contaminated compared to a village located at 50 km from the smelter. A study was carried out in Lagos city, Nigeria to compare metal pollution levels in vegetables and soils from industrial and residential areas (Yusuf *et al.* 2003). The study indicated that metal pollution was higher in industrial areas and discouraged vegetable planting in industrial areas.

Metal accumulation in plants depends on the plant species, types of soil, environment and agricultural practice (Alloway 1990). Some plants may accumulate toxic metals at levels which may be harmless to the plant but could be harmful to humans if ingested (Radojevic and Bashkin 1998). The total metal content in soil is the result of parent materials, fertilisers, atmospheric deposition, agrichemicals, and organic wastes (Alloway 1990). Metals like zinc, cobalt and copper are essential group of metals required for some metabolic activities (Awofolu *et al*. 2005). Lead and cadmium are very toxic and excessive content of these elements can lead to cardiovascular, kidney and nervous system diseases (WHO 1995).

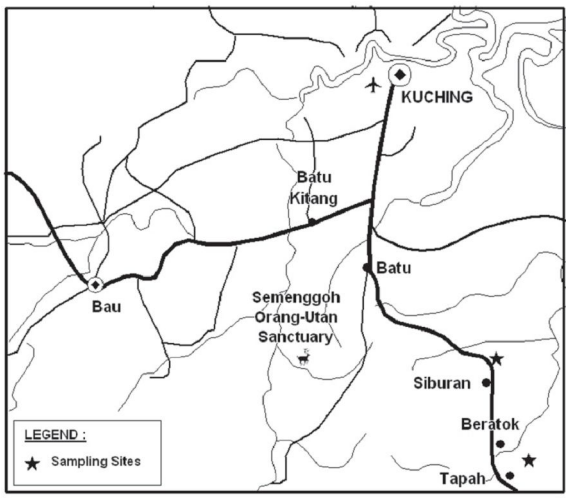
The Malaysian dietary guidelines recommend five servings of fruits and vegetables for adults to maintain good health (Khairiah *et al.* 2004). Largely, due to awareness of vegetable consumption, large quantities of vegetables are consumed to obtain sufficient nutrients as body supplements and this has resulted in human beings being exposed to risk from contaminated soils and vegetables. Hence, this study aimed to investigate heavy metal uptake in cultivated leafy and fruit vegetables from two farmlands in Siburan and Beratok. Beratok is located nearer to the roadside compared to Siburan which is located far from the roadside. Higher metal concentrations were anticipated to occur in Beratok. Metal analyses in the edible parts of vegetables were carried out. In order to have a clear picture of metal distribution in each edible portion of the vegetables, the leaves, stems and roots were also analysed separately. The characteristics of the soil samples from the farmlands were also determined. Attempts were made to relate the metal contents in both compartments and the findings were compared with the Malaysian Food Act 1983 and FAO/WHO guidelines.

MJ of Soil Science 057-069.pmd 58 08-Apr-08, 10:46 AM

MATERIALS AND METHODS

Sample Collection

Samples were taken from agricultural farms at Siburan and Beratok in Kuching, Sarawak (*Fig. 1*). These farmlands were chosen for this study mainly because the vegetables harvested from these farmlands are supplied to the residents in these areas and also marketed to the nearby markets for public consumption. Vegetable samples including kale, green mustard, white mustard, long bean and cucumber were collected from three plots and samples were harvested in a 1m x 1 m quadrant. Vegetable species that were collected at each area are shown in Table 1. These vegetables are commonly consumed by the locals and can be easily cooked. Topsoil samples at depth of 0-30 cm (n = 35) were also collected from these plots. Two composite soil samples which comprised four sub-samples were taken randomly at each plot. Control soil samples were collected from nearby plots which were not planted with vegetables.



X- Sampling site

Fig. 1: Location of Siburan and Beratok

TABLE 1
Vegetable species (n= 30) collected at Siburan and Beratok

Sampling area	Types	Common name	Scientific name
Beratok	Leafy	Kale	*Brassica alboglabra*
		Green mustard	*Brassica chinensis*
		White mustard	*Brassica rapa*
Siburan	Leafy	Green mustard	*Brassica chinensis*
		Mustard	*Brassica sp.*
	Fruit	Cucumber	*Cucumis sativus*
		Long bean	*Vigna sinensis*

MJ of Soil Science 057-069.pmd 59 08-Apr-08, 10:46 AM

TABLE 2
Recovery percentage obtained for dry ashing and wet digestion method

Method	Recovery percentage (%)						
	Fe	Zn	Co	Cu	Cd	Pb	Mn
Wet digestion	90.3	97.0	103.9	92.4	97.6	94.2	95.5
Dry ashing	77.2	90.1	87.5	85.6	79.8	86.7	93.1

Sample Preparation and Plant Analysis

The vegetables were washed with tap water followed by distilled water to eliminate attached soil particulates. Samples were divided for application of two types of analysis. Some samples were used for metal analysis of all the edible parts. For metal analysis in different parts of the vegetables, leaves, stems and roots were separated and sliced into smaller pieces. Subsequent to this, the samples were freeze-dried to reduce the risk of losing volatile elements and ground into powder using pestle and mortar. Two types of dissolution methods were applied to assess metals recovery in the samples, namely wet digestion and dry ashing. In the wet digestion method, 1 g of sample was digested with a mixture of $HClO_4$ and HNO_3, while for dry ashing 1 g of sample was placed in a crucible and heated at 450°C in a muffle furnace for 2 hours. The ash was dissolved in 5 mL of concentrated HCl solution. Recovery analysis was done based on amount recovered from the spiked concentrations. Table 2 represents the recovery percentage obtained for the dissolution methods. It can be seen that the wet digestion method produced better recovery results for the metals. Hence, this method was employed to test the samples. Metals, Fe, Co, Zn, Mn, Cu and Pb were analysed using Flame Atomic Absorption Spectrometry (FAAS) (Model Perkin – Elmer 3110).

Soil Analysis

Soil samples were analysed for organic matter content, pH, soil texture, total phosphorus and total Kjeldahl nitrogen (TKN). Total organic matter was determined using loss of ignition method at 550°C (APHA 1995), while soil pH was measured using 1:5 soil water ratio. Particle-size analysis was done based on Stoke's law using the pipette method (USDA 1984). Total Kjeldahl nitrogen was measured by employing the micro Kjeldahl procedure with sulfuric acid and digestion catalyst. Total phosphorus was determined by molybdenum blue method. For heavy metal analysis, 1 g of soil sample was digested with 10 mL HNO_3 and 3 mL of H_2O_2. For Cd, Zn, Cu, Mn, Co and Pb analysis, a supernatant was used with flame atomic absorption spectrometry (FAAS). All metal contents are expressed in mg kg^{-1} dry weight.

Statistical Analysis

Statistical analysis was performed using Two-Way ANOVA without replicate analysis to compare the parameters analysed and metal concentrations between

MJ of Soil Science 057-069.pmd 60 08-Apr-08, 10:46 AM

sampling sites and vegetables. A correlation study was done to investigate the existence of any relationships between metal concentration in vegetables with soils.

RESULTS AND DISCUSSION

Heavy Metals in Vegetables

The mean concentrations of metals found in the edible portion of the vegetable samples are presented in Table 3. Wide variations were observed in the accumulation of metals by the vegetable samples which represented the selectivity of plants towards metals and other contributing factors such as the locations of the farmlands and types of fertilisers applied to the vegetables. Generally, leafy type vegetables accumulated higher level of metals compared to fruit type and this was further proven by statistical analysis ($p<0.05$). A previous study has

TABLE 3
Mean concentrations of heavy metals in edible parts of vegetables at Beratok and Siburan

Area	Sample	Concentration (mg/kg)					
		Fe	Co	Zn	Mn	Cu	Pb
Beratok	Kale	88 (73.00 – 108.67)	3.44 (2.33-4.67)	53.78 (33.33-81.00)	13.78 (6.00-25.00)	9.78 (6.33-15.33)	1.73 (1.93-2.90)
	Green mustard	338.67 (75.33-421.67)	4.28 (2.33-6.67)	71.06 (47.67-139.00)	26.71 (12.00-40.00)	8.61 (3.33-14.33)	1.22 (1.50-2.37)
	White mustard	138.50 (70.67-209.00)	2.94 (2.33-3.67)	59.83 (28.00-92.33)	20.11 (12.67-33. 00)	6.38 (3.00-12.33)	1.49 (1.50-2.90)
Siburan	Green mustard	174.39 (50.33-303.33)	4.61 (3.33-6.33)	66.39 (47.67-90.33)	55.67 (21.33-106.67)	5.89 (4.00-10.33)	1.34 (1.27-2.30)
	Mustard	240 (58.33-569.33)	3.67 (3.00-5.00)	47.33 (26.33-91.33)	38.05 (9.00-80.3)	4.00 (2.33-8.67)	1.24 (1.57-2.13)
	Cucumber	62.5 (61.33-63.67)	3.83 (3.00-4.67)	33.67 (33.00-34.33)	18.67 (16.00-21.33)	5.00 (2.45-10.58)	1.40 (1.57-2.03)
	Long bean	72 (65.33-78.67)	3.67 (3.33-4.00)	48.84 (36.67-61.00)	59.17 (46.00-72.33)	7.84 (7.67-8.00)	1.15 (1.73-2.18)

Values in parentheses are minimum and maximum concentrations

MJ of Soil Science 057-069.pmd 61 08-Apr-08, 10:46 AM

shown that leafy vegetables accumulate more metals compared to other vegetables (Jinadasa *et al*. 1997).

Fe, Zn, Mn and Cu were found to be present in high concentrations. It is known that Fe, Zn, Cu and Mn are essential elements in plant nutrition. Fe indicated an elevated concentration in leafy vegetables at Beratok (88 - 338.67 mg/kg dry weight) and Siburan (174.39 to 240 mg/kg) compared to fruit vegetables (62.5 - 72 mg/kg at Siburan). The mean concentration of Mn varied between 13.78 – 26.71 mg/kg at Beratok and 18.67 - 59.17 mg/kg at Siburan. Green mustard and long bean at Siburan recorded the highest amounts of Mn. Mn plays a vital role in mucopolysaccharide metabolism and is also related to superoxide dismutase (Vandeecasteele and Block 1993). Greater accumulation of Cu was found in kale at Beratok (9.78 mg/kg). This could be due to translocation of Cu from roots to leaves in which micronutrients are required by plants for metabolism (Hopkins 1999).

In general, the least abundant metals in the vegetables were Pb and Co. The levels of Co in Siburan and Beratok were similar. The mean level of Pb in several vegetables of Beratok exceeded slightly the permitted level of the Malaysian Food Act 1983 for Pb which is 1.5 mg/kg. However, comparing the level of Pb with FAO/WHO guidelines (Pb, 2.0 mg/kg) the mean level of Pb complied with the standard. The potential source of Pb could be due to gas emission from traffic as the Beratok farmland is located near the road way. This finding was in accordance with a study done on leafy vegetables that were grown near the road side in Addis Ababa which was also found to contain a higher level of the Pb element (Itanna 2002). Nabulo *et al*. (2006) reported that Pb concentration in vegetable weed, *Amaranthus dubius* leaves, decreased with increasing distance from the road edge. This study concluded that the dominant pathway for Pb contamination was from atmospheric deposition. Sharma *et al.* (2006) studied the variations in metal concentrations during different seasons where Pb and Ni concentrations were higher than in the Indian standard in both summer and winter seasons in the edible portions of *Beta vulgaris.*

Figs. 2-4 show the concentrations of metals in different parts of vegetables for kale, green mustard and white mustard. Metals in different parts varied between the types of vegetables. This may be attributed to the variation in heavy metals uptake by different types of vegetables. High concentrations of essential elements (Fe, Co, Cu, Mn and Zn) and non-essential metal (Pb) were found in the leaves of kale at Beratok (*Fig. 2*). Pb showed a tendency to concentrate in leaves of the vegetables compared to other parts at Siburan and Beratok (*Figs. 3 and 5*). Metal levels in different parts of green mustard and white mustard grown at both places did not show any difference. Literature on metal distribution in different parts of vegetables is insufficient.

Concentrations of metals in the vegetables at Siburan and Beratok were influenced by the water used for irrigation, spraying methods and also the fertilisers

MJ of Soil Science 057-069.pmd 62 08-Apr-08, 10:46 AM

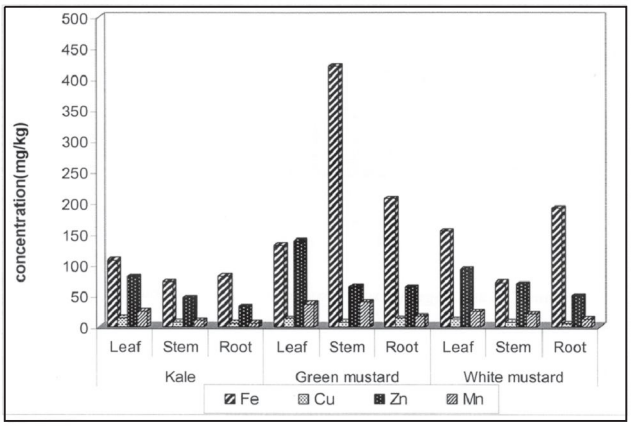


Fig. 2: Concentrations of Fe, Cu, Zn and Mn in different parts of vegetables at Beratok

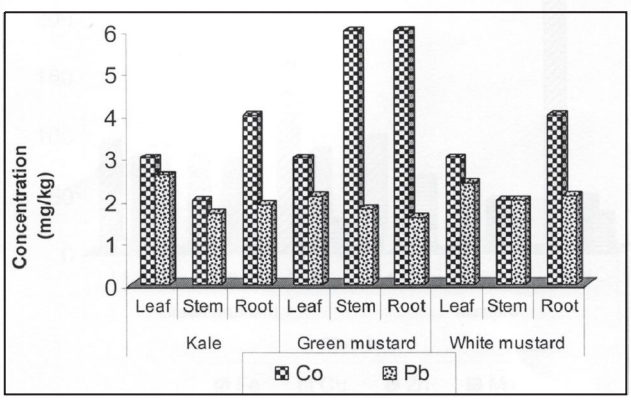


Fig. 3: Concentrations of Co and Pb in different parts of vegetables at Beratok

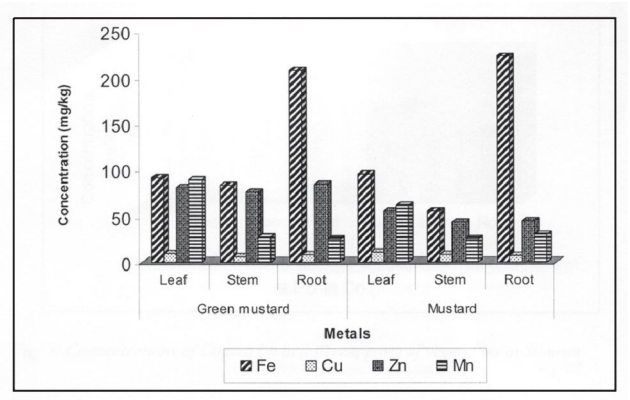


Fig. 4: Concentrations of Fe, Cu, Zn and Mn in different parts of vegetables at Siburan

MJ of Soil Science 057-069.pmd 63 08-Apr-08, 10:46 AM

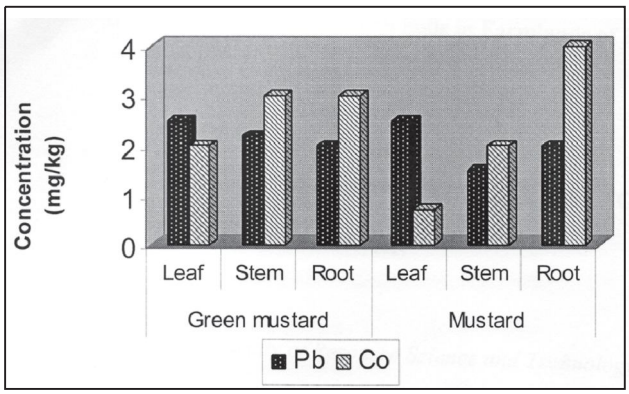


Fig. 5: Concentrations of Co and Pb in different parts of vegetables at Siburan

used. At Siburan, irrigation water from nearby Sungai(river) Ribit was applied to the vegetables. The accumulation effect depends strongly on the crop's physiological properties, the mobility of the metals, and the availability of metals in soils (Liu *et al.* 2005) and surface deposits on the parts of vegetables exposed to polluted air (Buchaver 1973).

Soil Properties

The physico-chemical properties of soils collected from the plots are tabulated in Table 4. The soil samples in Siburan and Beratok were loamy soils. The pH values of the soils from the plots of Siburan and Beratok ranged from 6.46 - 6.65 which reflect mild acidic soils. This was similar to soil pH values found in Linyi, Yntai and Xinxiang, China which ranged from 6.32 - 6.65 due to wastewater irrigation (Wang *et al.* 2004). Phosphatic fertilisers such as superphosphate can have considerable acidifying effect on soils. It is known that pH affects the bioavailability of soil metals but there were no major differences in pH between the sites. High rainfall during the sampling months could have lead to leaching, eventually resulting in low soil pH. However, the control plots were found to be moderately alkaline (pH 6.98 – 7.21) due to less wastewater irrigation presence and less usage of fertilisers and compost. It is known that the alkaline range of soils is known to restrict the mobilisation of heavy metals and thus reduce the uptake of heavy metals (Sharma *et al.* 2007).

Organic matter did not show major distinction between the plots and the areas. Soil texture, the content of sand, silt and clay showed differing fraction concentrations. Statistical analysis showed significant difference ($p<0.05$) in the means of sand, silt and clay fractions. The sand fraction was found to be dominant, followed by clay and silt. The movement of air and water to plant roots through soil is affected by soil texture. A good network of soil pores would allow rapid exchange of air and water with plant roots. Plant growth depends on rapid rates of exchange. Total phosphorus exhibited high levels in the soil samples. High amounts of phosphorus might be sourced from the use of compost as fertilisers in the farmlands. The percentage of total Kjeldahl nitrogen (TKN) is

MJ of Soil Science 057-069.pmd 64 08-Apr-08, 10:46 AM

TABLE 4
Soil characteristics of Beratok and Siburan

Parameters	Beratok				Siburan			
	Plot1	Plot2	Plot3	Control	Plot1	Plot2	Plot3	Control
pH	6.48	6.46	6.52	7.21	6.53	6.65	6.55	6.98
OM(%)	3.20	3.80	4.00	4.00	3.80	4.20	5.60	3.40
Clay (%)	36.4	32.4	35.5	27.8	41.9	40.8	40.4	51.8
Silt (%)	13.1	17.6	16.5	18.6	22.3	28.2	23.5	22.2
Sand (%)	50.5	50.0	48.0	53.6	35.8	31.0	36.1	26.0
Phosphorus (ppm)	193.2	123.2	159.2	188.7	108.4	176.5	163.3	137.1
Nitrogen(%)	0.16	0.17	0.20	0.17	0.15	0.12	0.20	0.10
Cd (mg/kg)	1.00	1.24	2.00	1.33	2.67	2.00	3.17	0.80
Co(mg/kg)	1.98	2.56	2.46	1.67	11.00	5.00	4.10	5.33
Zn(mg/kg)	66.67	63.33	66.58	85.45	76.70	63.33	78.35	93.50
Mn(mg/kg)	127.05	120.65	183.33	155.00	137.67	157.67	100.25	143.34
Cu(mg/kg)	123.34	146.70	126.80	126.70	120.52	123.36	147.85	60.00
Pb(mg/kg)	22.10	10.00	17.67	20.67	25.33	22.00	21.33	20.33

a reflection of of ammonia-nitrogen and organic nitrogen content in soils. Nitrogen content varied between 0.16-0.20 and 0.10-0.20% at Beratok and Siburan, respectively, indicating no differences.

Elevated concentrations of Mn and Cu were found in the soils. The concentrations of Cu and Mn were higher than in the vegetables species. Mn occurs naturally in rock, whereas Cu levels in soil could be affected by soil and crop treatment such as fungicides, fertilisers and also chicken dung (Alloway 1990). The levels of Cu obtained at Siburan and Beratok ranged from 120.52 – 147.85 mg/kg and 123.34 – 146.70 mg/kg, respectively. Similar levels of Cu was found in soils under vegetables in Harare, Zimbabwe which showed total concentrations of Cu ranging from 7.0 to 145 mg/kg (Mapanda 2005).

Comparably, the concentrations of Pb in soils were much higher than those found in the vegetables. The levels of Pb in Siburan and Beratok were 20.33-25.33 mg/kg and 10.00-22.10 mg/kg, respectively. The concentrations of Pb in uncontaminated soils were <20 mg/kg. Due to low solubility and relative freedom from microbial degradation, Pb tends to accumulate in soils and sediments and remain bioavailable. The maximum allowable limits (MAL) in soils suggested by Kabata-Pendias and Pendias (1992) are 5 mg/kg for Cd, 50 mg/kg for Co, 100 mg/kg for Cu and Pb and 300 mg/kg for Zn. The concentrations of Co, Zn, Cu and Pb in the soils of Siburan and Beratok were below the MAL and can be classified as uncontaminated soils.

A correlation study was carried out to ascertain if there is any relationship between Cu, Zn and Pb in vegetables and that of soils. A moderate correlation was observed between plant Pb and soil Pb ($r = 0.65$) at Siburan and Beratok. A

MJ of Soil Science 057-069.pmd 65 08-Apr-08, 10:46 AM

TABLE 5
Transfer coefficients of Cu, Zn and Pb from soil to vegetables

Area		Cu	Zn	Pb
Siburan	Kale	0.33	5.8	4.7
	Green mustard	0.27	3.6	7.5
	White mustard	0.08	5.4	6.8
Beratok	Green mustard	0.67	7.1	5.2
	Mustard	0.45	2.5	4.1
	Cucumber	0.35	4.8	5.5
	Long bean	0.60	7.5	4.6

significant degree of correlation was observed between Zn level in vegetables and Zn level in soils (r = 0.86; P=0.001) and a moderate correlation occurred between vegetable Cu and Cu in soil (r = 0.55). The soil metal concentrations appear to influence the uptake of Zn and to a lesser extent Cu. There have also been reports to show the relationship between metals in soils and vegetables during different seasons. Sharma *et al* (2006) concluded that Zn in soil had a positive significant relationship with vegetable contamination during winter, while concentrations of Cd, Cu, and Mn in soil and plant showed significant positive relationships only during summer.

Soil-plant Transfer Coefficients

The transfer coefficient is the metal concentration in plant tissue above the ground divided by the total metal concentration in the soil. Table 5 shows the transfer coefficients of metals at Siburan and Beratok. The transfer coefficients as suggested by Kloke *et al.* (1994) for Cu and Pb (0.01-0.1), and Zn (1-10) were used for comparison. Zn and Pb showed the highest transfer coefficients with no samples exceeding the Zn and Pb transfer coefficients limits. Samples exceeded the coefficient range suggested for Cu. This could have resulted from elevated concentrations of Cu in the soils.

Kachenko and Singh (2004) reported that soil properties may have influenced the soil-plant transfer of Cu because at Boolaroo (Pb-Zn smelter), 82% of samples exceeded the suggested coefficient range of Cu and 63% exceeded the suggested Pb coefficient range. The transfer coefficients differed between the vegetables species and sampling locations due to soil properties. Mohamed *et al* (2003) also reported different concentration factors (CF) from vegetables to vegetables and the magnitudes of the CF values reflect that the uptake of the major elements (K, Mg and Ca) are more pronounced than those of the trace elements (Co, Cu, Fe, Ni and Zn).

MJ of Soil Science 057-069.pmd 66 08-Apr-08, 10:46 AM

CONCLUSION

The assessment of metals in vegetables showed that leafy vegetables such as kale, white mustard and green mustard exhibited higher element concentrations than the fruit types. The toxic element, Pb was found in the least concentrations compared to other metals analysed. However, the level of Pb exceeded the Malaysian Food Act 1983 guidelines. The levels of metals in similar vegetable species differed between the sampling sites at Siburan and Beratok. It is suggested that soil properties between the sampling sites contributed to this variation.

There are various sources that could attribute to metal contamination and contamination process which could have taken place during pre-harvest and post harvest process. Sources during pre-harvest include usage of compost, fertilisers and pesticides, chicken dung and water used for irrigation, while sources during post harvest include contamination through air and also during transport to market. Future research should consider the effects of irrigation water and compost or fertilisers on the availability of metals to different species of vegetables and soils. Microbiological studies also should be undertaken to evaluate the presence of pathogens.

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MJ of Soil Science 057-069.pmd 68 08-Apr-08, 10:46 AM

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MJ of Soil Science 057-069.pmd 69 08-Apr-08, 10:46 AM