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Research Article

Heavy Metal Content in Soils and Vegetables Grown in an Inland Valley of Terengganu and a River Delta of Kelantan, Malaysia

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Abstract: Heavy metal concentration in vegetables grown in an inland valley of Terengganu and a deltaic area of Kelantan are compared. The presence of heavy metals as indicated by speciation of Fe, Mn, Pb, Cr, Cu, Zn and Cd are discussed. Vegetable farm soils in Terengganu and Kelantan are acidic, sandy and contain a fair amount of organic carbon. Most of the soil Fe is highly concentrated in the RR fraction in the form of mottles of iron oxide and coatings on the sediment surface. Manganese is highly concentrated in the AR fraction, followed by its occurrence in the OO fraction. The study showed the presence of precipitates of Mn-oxyhydroxides and the association of Mn with soil organic carbon. The rest of the heavy metals (Pb, Cr, Cu, Zn and Cd) studied were significantly concentrated in the OO fraction suggesting the great affinity of these metals to the organic carbon in the soils. The fair amount of organic carbon present in the study area might possibly be attributed to litter constituting plant materials and poultry manure-amended soils in the vegetable farms. Despite the low concentration of Pb in the available form (ELFE fraction), its accumulation in the plants could have occurred because of the Pb-organic complexes in the soil. The accumulation of Cd in the soils could be attributed to the presence of available Cd (ELFE fraction) in the soils.

Keywords: Heavy metal, river delta, soils, vegetables

INTRODUCTION

Vegetables are the third largest crop grown in Malaysia after the cash crops such as palm oil and rubber and the staple food crop, paddy. The acreage of vegetable farms in Malaysia is currently 53,322 ha with a total production of 878,975 tons (Anon, 2012). Most vegetable areas are located at areas secondary to the cash crop areas. According to Tee (1979) vegetables were previously only grown in small patches by the rural folk for their own consumption and the bulk of the urban needs were imported.

However, urbanization and rapid industrial development stimulated the demand for fresh greens in urban centers. Land in the outskirts of town centers were converted into vegetable farms that were mainly operated by Chinese farmers. Larger farms were concentrated in specialized areas in the highlands to produce temperate vegetables, while the peat regions of Johore were utilized for growing fresh vegetables for export to Singapore. Cameron Highlands had one of the earliest commercial vegetable farms in Malaysia (Ding *et al*., 1981). Collectively, thousands of farms scattered all over the country produce fresh vegetables throughout the year (Tee, 1979).

High temperature, humidity, heavy rainfall and sunshine throughout the year offer a conducive environment for a continuous cycle of pests and diseases. To overcome this problem and to meet the local demand for vegetables, various types of pesticides and fertilizers have been used. Previous research has demonstrated that the application of chemical fertilizers and pesticides increased the heavy metal content in the soils and vegetables. Besides that, other sources of pollution that contribute to the accumulation of heavy metals in agricultural soils are polluted air and the use of sewage sludge as fertilizer (Zhang *et al*., 1997). The fungicide chlorothalonil and insecticides including organophosphates, chlorpyrifos and profenofos are widely used in vegetable farms (Ngan *et al*., 2005). The pesticides are applied intensively because the high humidity and cultivation throughout the year are conducive to pest growth and hence cause infestation of the vegetable crops (Hanada, 1987).

The bioavailability of elements or heavy metals to plants is subject to many factors associated with soil and climatic conditions, plant genotype and agronomic management, including active/passive transfer process, sequestration and speciation, redox condition, the plant

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root system type and the plant's response to elements in relation to seasonal cycles (Kabata-Pendias and Pendias, 1984). Toxicity or necessity varies from element to element and from species to species (Tripati *et al*., 1997).

The aim of the present study was to investigate the availability of heavy metals in the soils and in the vegetables grown at selected agricultural areas in Terengganu and Kelantan which represent two different types of vegetables soils. The study area in Kelantan was a deltaic area whereas the soil in Terengganu was developed on an inland valley. These above mentioned areas have slightly different lithology that could have influenced content and the availability of heavy metals, in the respective soils. The sequential extraction method was adopted to determine the availability of metals from the soil to the plant and the potential of the metals in the soil to leach out (from the two different types of soil). Previous studies have indicated that elements or metals exist in the soil in different forms depending on their origin as well as their chemical and physical characteristics.

Not all metals in the soil have the potential to be taken up by plants. Only those in the available form are easily absorbed by the root system of the plant through nutrient uptake and thus are able to enter the food chain and later pose a risk to human life. Hence the present study is very useful because the potential availability and mobility of heavy metals in the soils is investigated for two soil types (which represent inland valley and deltaic soils) from Terengganu and Kelantan.

MATERIALS AND METHODS

Site location: Three sites at Marang, Kuala Berang and Kuala Terengganu in Terengganu state and two areas at Tumpat and Kelaboran in Kelantan state were selected for the study. Both states are located at the northeastern part of Peninsular Malaysia. Most of the areas comprised small vegetable farms run by local farmers growing vegetables for the local market. Vegetables selected for the study were leafy vegetables which are commonly consumed by Malaysians namely spinach mustard (*Brassica juncea*) or sawi pahit in Malay, amaranth (*Amaranthus gangeticus* L.) or bayam in Malay and water convolvulus (*Ipomoea aquatica* Forsk.) or kangkung in Malay. For plant sampling, the whole plant was uprooted and soil samples were taken from 0 to 30 cm depth. All samples were placed in separate plastic bags and transported to the laboratory for further analysis.

Vegetable samples: The vegetable samples were washed initially in running water to remove soil particles followed by washing with distilled water and dionized-distilled water. The samples were dried with clean tissue before being cut into small pieces and separated into leaf, stem and root parts. The samples were then oven dried at 70°C to constant weight. The dried samples were pulverized with a mortar and pestle and subjected to wet digestion in conical flasks containing HNO_3: $HClO_4$ (2:1) for 3 h on a sand bath (AOAC, 1984). The digested samples were filtered through Milliphore filter paper (0.45 µm pore size).

Soil samples: Soil samples were air-dried in the laboratory environment, ground separately using a mortar and pestle and then sieved through a 250 mm sieve prior to the metal extraction process. The sequential extraction method was adopted for the four soil fractions according to that of Badri and Aston (1983). From the first fraction, called the easily leachable and ion exchange (ELFE) fraction, metals from easily and loosely bonded compounds in the soil were extracted. For this method, 10 g of soil were weighed into a kartell bottle, followed by the addition of 50 mL of 1.0 M NH_4CH_3OO (pH7). The samples were then shaken for 1 ½ h before being centrifuged for ½ h at 3000 rpm. This was followed by filtering using Millipore filter paper of 0.45 µm pore size. For washing, 50 mL of distilled, dionised water was added followed by the mixture being shaken and centrifuged in the manner as described earlier. For the second fraction, the Acid Reducible fraction (AR) 50 mL of NH_2OH. HCl (pH2) was added into the kartell bottle followed by the same process of shaking, centrifuging and washing.

For the third fraction, the Organic Oxidation fraction (OO), containing metals bonded strongly to organic matter, the extraction process was done by adding 15 mL of H_2O_2 into the kartell bottle which was then placed in boiling water for 1-1 ½ h. Then 50 mL of NH_4CH_3OO was added, followed by same procedure as mentioned above. For the fourth fraction, the Resistant Fraction (RR) metals were extracted from silicate minerals by means of a wet digestion with HNO_3: $HClO_4$ (25:10) in a conical flask on a sand bath. This procedure was repeated until the sample turned whitish and this was followed by the filtration process.

Determination of heavy metals in all the studied samples were performed using the ICP-MS ELAN 9000 (Perkin Elmer model). The physico-chemical characteristics of the soil from the studied areas are given in Table 1. The data includes organic matter content (Walkley and Black, 1934), soil pH (Dudridge and Wainright, 1981) and grain size (Badri, 1984). The soil organic matter was slightly higher in the soil samples from Terengganu, while the pH of the Terengganu soil was lower than that of the soil from Kelantan with the exception of soil grown with spinach mustard.

RESULTS AND DISCUSSION

Heavy metal content in the studied vegetables: Heavy metal concentration in the studied vegetables

Table 1: The average of pH, organic carbon content and grain size of the soil from the study areas

Vegetable type	Soil pH	The percentage of organic carbon	The percentage of grain size
Terengganu Spinach mustard	6.03±0.02[a]	2.56±0.1[a]	32.4±3.304[a]
Amaranth	4.23±0.05[b]	2.46±0.1[b]	36.76±2.577[b]
Water convolvulus	4.13±0.15[c]	2.13±0.2[c]	43.86±2.49[c]
Average	4.796±0.007	2.38±0.13	37.673±2.79
Kelantan Spinach mustard	5.771±0.124[a]	1.779±0.095[a]	13.666±1.741[a]
Amaranth	5.548±0.264[b]	1.649±0.055[b]	14.583±1.727[b]
Water convolvulus	5.928±0.047[c]	1.919±0.14[c]	18.333±1.521[c]
Average	5.749±0.145	1.782±0.096	15.527±1.663

Means within the same column followed by the same letter are not significantly different from each other at $p<0.05$

Table 2: Range of heavy metals concentration in selected vegetables from Terengganu and Kelantan (mg/kg)

Species	Pb	Cd	Cu	Zn	Fe	Mn
Terengganu Spinach mustard						
Leaf	0.016-0.064	0.003-0.007	0.257-0.367	3.607-3.918	21.191-27.878	3.071-3.608
Stem	0.009-0.044	0.002-0.003	0.126-0.245	2.948-3.452	7.153-7.919	1.670-1.835
Root	0.110-0.135	0.008-0.010	0.008-0.010	5.371-8.205	76.026-161.660	4.983-5.124
Amaranth						
Leaf	0.021-0.028	0.004-0.007	0.844-0.908	3.313-6.419	45.860-46.929	5.167-5.506
Stem	0.009-0.012	0.004-0.006	0.211-0.235	2.129-2.947	7.775-12.441	1.760-5.490
Root	0.077-0.164	0.008-0.045	0.177-0.408	3.846-6.914	53.001-114.939	19.682-33.339
Water convolvulus						
Leaf	0.025-0.043	0.005-0.007	0.201-1.161	3.985-4.128	25.840-30.400	15.467-16.947
Stem	0.016-0.017	0.003-0.009	0.055-0.628	2.465-3.667	10.326-12.414	3.907-6.998
Root	0.150-0.710	0.016-0.033	1.164-1.224	7.726-12.367	80.651-226.946	19.464-34.468
Kelantan Spinach mustard						
Leaf	0.026-0.035	0.004-0.007	1.059-2.033	6.268-8.268	25.550-33.626	4.900-5.729
Stem	0.008-0.010	0.001-0.004	0.333-1.554	2.461-3.769	7.713-10.716	1.872-2.719
Root	0.067-0.097	0.005-0.014	2.679-5.519	8.781-11.795	47.030-66.487	7.152-10.612
Amaranth						
Leaf	0.048-0.057	0.012-0.027	1.388-1.580	8.844-11.717	36.599-56.522	12.885-13.608
Stem	0.006-0.014	0.002-0.009	0.271-0.717	2.945-6.198	13.938-16.073	6.943-8.900
Root	0.053-0.066	0.014-0.029	1.521-1.607	17.487-18.202	59.738-61.378	26.937-33.908
Water convolvulus						
Leaf	0.026-0.026	0.004-0.005	0.435-0.976	3.683-7.220	21.792-23.345	5.537-9.356
Stem	0.009-0.010	0.003-0.004	0.173-0.294	1.418-3.288	7.999-9.544	2.429-5.303
Root	0.058-0.110	0.010-0.022	1.724-3.771	11.213-15.662	32.742-65.008	25.293-27.919

from the sampling sites in Terengganu and Kelantan are shown in Table 2. In general toxic metals such as Pb and Cd were present, but in small quantities. Both these metal had the highest content in the root followed by the leaf and stem.

Concentration levels of both the toxic metals (Pb, Cd) in the study areas were comparable to those obtained in a previous study on selected areas in Malaysia (Khairiah *et al*., 2003, 2004; Ismail *et al*., 2005). According to Guo *et al*. (2006) leafy vegetables such as Chinese cabbage or dwarf white mustard (*Brassica chinensis* L) and water convolvulus (*I. aquatic*) in southeastern China, accumulate Cd and Pb more easily than non-leafy vegetables. On the other hand past research also showed that Cd was more easily taken up and distributed to all plant parts, compared to Pb. The present study also indicated the same result where low concentrations of Cd and Pb were detected in the studied vegetables from both areas.

The other metals such as Cu, Zn, Fe and Mn were expected to be present in relatively high amounts in all the studied vegetables due to their involvement in various metabolic activities in the plant (Hopkins, 1999).

Heavy metal content in the soil: Vegetable farm soils of Terengganu were developed on alluvial deposits in the inland valley of the Terengganu river. This valley was overlain by levees, cut-off meanders and abandoned canal deposits. Lithologically the deposits ranged from poorly-sorted silty sand to sandy silt (Bosch, 1988). In the study, the nature of the soils were sandy (with 32.4-43.86% grain size <63 μm), acidic (pH 4.13-6.03) and contained a fair amount of organic carbon (2.13-2.56%). In Kelantan, the sampling sites were located at the southern part of the Kelantan river delta. The soils were developed on the inner beach ridges containing sand, clay and silt (Bosch, 1988). It was found that the vegetable farm soils in Kelantan were sandy (with 13.66-18.33% grain size <63 μm), had less amount of organic carbon (1.65-1.92%) and were slightly acidic (pH 5.55-5.92).

In general, the total concentration of Fe, Mn, Cd and Cr in the soils of Terengganu and Kelantan were similar. The Pb concentration in the Terengganu soils was higher than that of the Kelantan soils. However, the concentration of Cu and Zn in the Terengganu soils was lower than that of the Kelantan soils.

Except for Fe and Mn, all of the heavy metals studied tended to accumulate in the OO fraction,

Table 3: Fe, Mn and Pb speciation in vegetables farm soils of Terengganu and Kelantan

Heavy metals speciation (mg/kg)	Terengganu			Kelantan	
	Marang	Kuala Berang	Kuala Terengganu	Tumpat	Kelaboran
Fe					
ELFE	10.21±7.09[b]	10.07±3.23[c]	3.90±0.89[c]	4.47±3.27[b]	6.01±2.88[b]
AR	59.78±29.20[b]	39.77±25.11[c]	74.38±18.18[b]	36.56±3.22[b]	40.76±4.20[b]
OO	66.03±18.99[b]	89.87±11.27[b]	52.08±3.18[b]	36.72±11.82[b]	41.45±11.42[b]
RR	324.83±148.39[a]	158.85±48.2[a]	188.39±29.08[a]	241.91±80.49[a]	279.54±110.86[a]
Total	460.86	298.56	318.75	319.66	367.75
Mn					
ELFE	2.61±1.22[b]	4.26±0.89[b]	1.17±0.52[c]	2.09±1.21[c]	1.75±0.88[c]
AR	7.40±4.59[a]	15.10±4.76[a]	3.87±0.41[a]	9.83±1.23[a]	14.80±3.96[a]
OO	7.88±3.47[a]	13.35±5.26[a]	3.14±0.78[ab]	4.38±1.71[b]	5.13±2.62[b]
RR	7.64±4.99[a]	8.41±3.05 [b]	2.61±0.22[b]	1.03±0.60[c]	1.37±0.58[c]
Total	25.54	41.12	10.79	17.34	23.05
Pb					
ELFE	0.10±0.04[c]	0.07±0.03[b]	0.02±0.01[c]	0.10±0.04[c]	0.10±0.09[c]
AR	0.22±0.13[c]	0.22±0.13[b]	0.08±0.02[c]	0.05±0.02[c]	0.08±0.07[c]
OO	4.28±1.61[a]	5.39±3.28[a]	4.73±1.20[a]	1.19±0.71[a]	1.28±0.68[a]
RR	2.75±1.04[b]	4.56±0.59[a]	2.67±0.40[b]	0.50±0.27[b]	0.72±0.34[b]
Total	7.35	10.25	7.51	1.84	2.18

Means within the same column followed by the same alphabet are not significantly different from each other at $p<0.05$ in the same area

Table 4: Cd, Cu, Cr and Zn speciation in vegetables farm soils of Terengganu and Kelantan

Heavy metals speciation (mg/kg)	Terengganu			Kelantan	
	Marang	Kuala Berang	Kuala Terengganu	Tumpat	Kelaboran
Cd					
ELFE	0.01±0.01[ab]	0.02±0.01[b]	ND	0.01±0.00[bc]	0.01±0.01[b]
AR	0.01±0.00[b]	0.02±0.01[b]	ND	0.01±0.00[b]	0.02±0.01[b]
OO	0.02±0.00[a]	0.03±0.02[a]	0.01±0.00[a]	0.02±0.00[a]	0.03±0.02[a]
RR	ND	ND	ND	0.01±0.00[c]	0.01±0.01[b]
Total	0.04	0.07	0.01	0.05	0.07
Cu					
ELFE	0.12±0.06[c]	0.19±0.06[b]	0.13±0.17[b]	0.21±0.06[c]	0.34±0.04[c]
AR	0.04±0.02[c]	0.14±0.08[b]	0.01±0.00[b]	0.10±0.03[c]	0.19±0.07[c]
OO	4.31±0.99[a]	6.42±2.38[a]	1.48±0.40[a]	7.66±1.01[a]	8.94±2.20[a]
RR	0.75±0.27[b]	1.02±0.21[b]	0.40±0.00[b]	1.90±0.71[b]	3.12±1.30[b]
Total	5.23	7.78	2.01	9.88	12.58
Cr					
ELFE	0.29±0.05[c]	0.29±0.03[c]	0.27±0.01[b]	0.28±0.05[b]	0.27±0.04[c]
AR	0.05±0.00[c]	0.05±0.00[c]	0.05±0.00[b]	0.06±0.01[b]	0.07±0.00[c]
OO	3.56±1.54[a]	3.98±1.09[a]	2.70±0.87[a]	2.54±1.12[a]	4.73±1.26[a]
RR	1.25±0.59[b]	1.49±0.36[b]	0.76±0.08[b]	0.62±0.33[b]	1.16±0.51[b]
Total	5.15	5.82	3.79	3.50	6.23
Zn					
ELFE	0.74±0.43[c]	0.94±0.35[b]	0.56±0.05[b]	1.59±0.77[c]	1.81±0.88[bc]
AR	0.93±0.34[c]	2.07±0.94[b]	1.12±0.97[b]	2.76±1.17[b]	3.81±1.64[b]
OO	4.08±1.07[a]	8.68±3.96[a]	6.24±2.94[a]	5.48±1.72[a]	6.86±3.89[a]
RR	1.83±0.44[b]	2.80±0.50[b]	1.54±0.13[b]	1.08±0.51[c]	1.29±0.42[c]
Total	7.58	14.49	9.46	10.90	13.77

Means within the same column followed by the same alphabet are not significantly different to each other at $p<0.05$ in the same area; ND: Not Detected

indicating the high association of Pb, Cd, Cr, Cu and Zn with organic matter Table 3 and 4. According to Anizan (1992), sandy soils from the east coast of Peninsular Malaysia have a monomorphic organic coating with varying thickness and colour. The reddish colour was caused by the high amount of iron in the amorphous organic matter. A high amount of iron nodules was also observed in the soils. The fair amount of organic carbon in the soils could be attributed to the poultry manure used to increase yields. According to Aini (2006), processed chicken manure has been used widely as organic fertilizer in vegetable farms due to its high content of N, P and K besides Ca. These are essential nutrients required for vegetable cultivation. Vimala *et al*. (2006) found that the processed poultry manure contained a high quantity of Fe followed by Zn and Mn. Vegetable farmers were encouraged to use Processed Poultry Manure (PPM) because it was cheap and easily available compared to chemical fertilizers plus it has been found to increase the yield of many crops in Malaysia. The application of PPM also resulted in the improvement of the chemical properties of the soil.

Soil Pb was mainly concentrated in the OO fraction (associated with organic matter), followed by the RR fraction (in a resistant form). Despite its low concentration in the available form (ELFE fraction), Pb was detected in the vegetable roots, leaf and stem parts. According to Kabata-Pendias and Pendias (1984), although Pb was not readily soluble in the soil, it was absorbed mainly by the root hairs and was stored to a considerable degree in the cell walls. There was also an alternative suggestion that Pb was not taken up directly from the soil by the plant roots, but rather was sorbed from dead plant materials that accumulated near the soil surface. In the present study, vegetables could have absorbed this metal from Pb-organic complexes in the soils.

In the study areas, Cd was highly concentrated in the OO fraction, indicating the good association between Cd and organic matter. The concentration of Cd was low in the Terengganu soils and was not detected in a few fractions. Although it was not detected in the RR fraction, soil Cd in Marang was highly concentrated in the OO and ELFE fractions. This metal was also detected in the vegetable root, leaf and stem parts. According to Kabata-Pendias and Pendias (1984), the soluble form of Cd in the soil was always easily available to plants. Cd can also be transported within a plant in the form of metalo-organic complexes. Cd was localized mainly in the roots, with lesser amounts in the stem nodes, petioles and major leaf veins. In the study, the accumulation of Cd in the plants could be attributed to the ELFE Cd in the studied soils.

The concentration of Fe in all the soil samples studied was significantly high in the RR fraction, followed by the OO and AR fractions. These results are in agreement with reports from other agricultural areas in Peninsular Malaysia (Khairiah *et al*., 2009, 2012). Soils at the deltaic (Kelantan) and inland valley (Terengganu) areas were highly exposed to oxidizing conditions. The soils were occasionally submerged because of the shallow water table during the rainy season and exposed during the dry season. Under these conditions most of the Fe would occur as resistant iron oxide mottles and coatings on the sediment surface (Kämpf *et al*., 2000). In such areas, the concentration of available Fe in the soil was low.

In the Kelantan soils, Mn was significantly high in the AR fraction whereas in the Terengganu soils, the Mn concentration was high in the AR and OO fractions. In the areas of seasonal water table fluctuation due to the rainy and dry seasons, plus the presence of interfering organic and inorganic components in the soil solution, fine grain size and poor crystallinity of Mn oxides occurred (Kämpf *et al*., 2000). In the sandy soils of Kelantan, the precipitates of Mn oxides could be found on the sediment surface. The soils of Terengganu were less sandy and had slightly higher percentage organic carbon than those of Kelantan. It is suggested that, apart from precipitates of Mn oxides, the soil Mn in the Terengganu soils was also highly associated with the organic matter.

CONCLUSION

Pb and Cd mainly accumulated in the roots, followed by that in the leaf and stem parts. In sandy soils of the Kelantan delta and inland valley soils of the Terengganu river, most of the heavy metals (Pb, Cr, Cu, Cd, Zn) studied were closely associated with the organic matter. The fair amount of organic matter in the soils could be attributed to litter as well as poultry manure amended soils in the vegetables farms. The low amount of available Pb in the soils, could have come from the Pb-organic complexes. The accumulation of Cd in the plants could be attributed to the available Cd in the ELFE fraction of the soils. Most of the soil Fe occurred as resistant iron oxides whereas Mn mainly occurred as precipitates of Mn oxides.

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