ORIGINAL ARTICLE

Accumulation and Health Risk of Heavy Metals in Cabbage Due to Long-term Mineral Fertilization From Vegetable Production Systems in Kundasang, Sabah

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ABSTRACT

Introduction: Fertilizers are widely applied in Malaysian agricultural areas to boost the growth of crops for better production. However, the repeated application of inorganic fertilizers has been shown to increase the metal concentration in soils due to impurities and background contamination. **Methods:** The concentrations of selected metals in cabbage (*Brassica oleracea var. capitata*) and soil sampled from the Monteki agricultural area of Sabah were investigated using flame atomic absorption spectrometry. **Results:** Concentrations of metals measured were in the order of zinc > copper > lead > cadmium, whereby none of the cabbage samples exceeded the concentration standards enumerated in the Malaysian Standards. Cabbage and soil samples from newer farms were characterised by significantly lower (P<0.05) concentrations of cadmium, copper, lead and zinc compared to older farms. A health risk analysis yielded target hazard quotient values <1 for individual metals in cabbage. The Hazard Index measuring the metal accumulation of risk through ingestion of cabbage was 0.081 and 0.763 for new and old farms, respectively. **Conclusion:** It can be concluded that there is no potential risk due to metal toxicity by ingestion of cabbage from this location.

Keywords: Metals, Cabbage, Hazard quotient, Risk estimation

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INTRODUCTION

A varied range of tropical and temperate vegetables are cultivated in Malaysia, supplying important sources of minerals and vitamins for diet requirements. Vegetables were originally grown in small plots for individual family consumption with the bulk of urban requirements being imported. Currently, a large amount of vegetables are grown in Malaysia for our own domestic use, with a total area of vegetable farms approximating 53,322 ha corresponding to a total production of 878,975 tons (1). Pesticides and fertilizers are extensively used in agricultural activities in Malaysia, to control crop pests and improve growth for better yields. The use of inorganic fertilizers may be good for the growth of crops but excessive use may result in potential sources of heavy metals such as arsenic, cadmium, copper, lead, mercury, nickel and zinc (2).

In Malaysia, the main contributor of heavy metals in agricultural areas is from consistent use of fertilizers (3). Previous studies have shown accumulation of heavy metals in soil and vegetables from agricultural areas in Sepang, Bangi, Cameron Highlands and Felda Tasik Chini (4). Heavy metals have the potential to build up in the soil when chemical fertilizers are used consistently causing soils to become contaminated thus enabling the crops grown on these soils to accumulate these contaminants (5). Repeated use of fertilizers has a substantial effect on metal concentrations and other soil properties such as pH, soil organic carbon, and cation exchange capacity, therefore greatly impacting metal availability (2,6).

The amount of metals in soil can contribute to the amount of metals accumulated in vegetables (7). Comparing leafy vegetable, fruit vegetable and root vegetable, accumulation of metals is much higher in leafy vegetables than the other two types of vegetables (8) due to the much higher translocation and transpiration rate in leafy vegetable types (9). Cabbage has been shown to have a high ability of absorbing heavy metal that have accumulated in soil (10). The translocation and uptake of metals into the edible portions of vegetables is a cause for concern due the risk of human exposure via consumption (11).

There is a high possibility of experiencing long term effects when humans are exposed to heavy metals. This is due to their property which is not biodegradable, therefore heavy metals that are not excreted from the body will accumulate in human organs causing various types of diseases depending on the type of element and severity of exposure (12). Documented effects on the human body due to heavy metal exposure include nerve tissue damage, liver damage, high blood pressure, darkening of the skin, paralysis, kidney damage, lung impairment and brittle bones (13). Approximately 66.28% of Malaysians reported eating cabbage in their weekly consumption of vegetables (14), therefore in this study we aimed to quantify the concentration of cadmium, copper, lead and zinc in soil and cabbage pertaining to the evaluation of potential human health risks.

MATERIALS AND METHODS

Field sampling

Specimens of soil and cabbage were taken from the Monteki Agricultural area in Kundasang, Sabah. The 8 acre sampling site was split into 4 subplots spanning 2 acres each whereby 10 samples were collected from each subplot. Soil and cabbage samples were taken from the same locality. Soil samples were collected within 15 cm from the surface in the root zone of the cabbage crops (15). Both samples were retained in suitable bags and sealed prior to being transported to the laboratory for analysis.

Sample analysis

Soil samples were dried at 105^{0}C in the oven for a minimum of 24 hours preceding acid digestion (16). The edible parts of the cabbage samples were first homogenized in a blender then dried at 105oC in an oven. Ten milliliters of concentrated nitric acid was added to 2 grams of dried sample (either soil or cabbage) and this mixture was heated at 60^{0}C for half an hour. After cooling, 2 mL of hydrogen peroxide was added and the solution was heated again until the solution became colourless with a volume of less than 5 mL. The digested samples were cooled and distilled water was added until the solution made a volume of 100 mL. Digested samples were analysed for metal concentration using the Perkin Elmer AAnalyst 400 AA Spectrometer.

Chronic Daily Intake of Metals

The chronic daily intake (CDI) of metals through consumption of cabbage was calculated based on the average cabbage consumption rate of 92g /day (17) and average body weight of 60 kg, whilst C is the metal concentration in cabbage calculated as µg/g.

$$CDI = \frac{IR \times C}{BW} \quad \text{Eq.1}$$

Health Risk Assessment

The potential health risk from dietary exposure to metals in cabbage among consumers was calculated using the following equation (18) to determine the Target Hazard Quotient:

$$THQ = \frac{EFr \times ED \times IR \times C}{RfD \times BW \times AT} \times 10^{-3} \quad \text{Eq. 2}$$

The values for the variables are as follows, Exposure Frequency = 365 days/year ; Exposure Duration = 70 years; and Averaging Time for non-carcinogens = 365 days/year Ч 30 years. The reference dose used was 0.001, 0.04, 0.004 and 0.3 mg/kg/d for cadmium, copper, lead and zinc, respectively (18). The Hazard Index (HI) was calculated by the summation of each metal THQ (19).

Statistical Analysis

The t-test was used to compare the means in the concentration of heavy metals in the soil between old and new farms. The relationship between metal concentrations in soil and cabbage was determined using the Pearson's correlation.

RESULTS

Metal concentration in soil

Forty samples of soil (n=40) were taken for the analysis, where the samples were equally divided among old and new farms. The areas classified as old farm had been in operation for more than 25 years while new farm was areas in operation for less than 25 years. The concentration of cadmium, copper, lead and zinc in soil samples is given in Table 1. All metals analyzed were detected in all sampling locations with a range of between 0 – 0.25 mg/kg, 8.55 – 40.15 mg/kg, 0.40 – 1.15 mg/kg and 21.5 – 100.5 mg/kg for cadmium, copper, lead and zinc, respectively. Cadmium was not detected in samples from both the new and old farms, whereas zinc was detected in the highest concentration. There was no significant difference in the cadmium concentrations between new and old farms ($p>0.586$). It was found that soil from new farms had significantly lower ($p< 0.01$) concentrations of copper, lead and zinc compared to soil from old farms (Table 1).

Metal concentration in cabbage and correlation with soil concentration

The results in Table 2 show the concentrations (mg/kg) of metals in cabbage samples. Zinc recorded the highest

Table 1: Soil concentration of different elements

Elements	Concentration (mg/kg)								*P* value
	New Farm (n = 20)				Old Farm (n = 20)				
	Mean	SD	Min	Max	Mean	SD	Min	Max	
Cd	0. 04	0.06	ND	0.15	0.06	0.07	ND	0.25	> 0.05
Cu	15.17	5.29	8.60	15.58	27.96	8.12	10.15	40.15	< 0.01*
Pb	0.60	0.15	0.40	0.85	1.00	0.22	0.45	1.35	< 0.01*
Zn	41.19	13.69	21.50	67.50	79.54	13.01	54.0	100.5	< 0.01*

ND non-detected, *P<0.01, statistically significant difference

concentration (43.5 mg/kg) while lead was not detected in all samples. It was found that cabbage grown on new farms had significantly lower concentrations of copper ($p < 0.01$), lead ($p < 0.05$) and zinc ($p < 0.01$) compared to cabbage sampled from old farms (Table 2). The mean concentration of copper, lead and zinc from old farms were 15.82 ± 3.9 mg/kg, 0.04 ± 0.07 mg/kg and 27.78 ± 7.96 mg/kg, respectively. All samples were below the permissible level for lead concentration according to the Malaysian Food Regulations 1985 (20) standard where the limit for lead is set at 2 mg/kg. There are no local standards for copper and zinc in vegetables, therefore guidelines from the World Health Organisation are used which proposed limits of 30 mg/kg and 50 mg/kg (21). The concentrations in the sampled cabbage were lower than the reported health regulatory limits for ingested vegetables.

Table 2: Element concentration in cabbage

Element	Concentration (mg/kg)				*P* value	^ MFR 1985 (mg/kg)	^^ WHO 2007 (mg/kg)
	New Farm		Old Farm				
	Mean	SD	Mean	SD			
Cu	7.16	2.59	15.82	3.90	< 0.01*		30
Pb	0.002	0.01	0.04	0.07	< 0.05**	2.0	
Zn	15.93	5.17	27.78	7.96	< 0.01*		50

* statistically significant at p < 0.01, ** statistically significant at p < 0.05
^ Malaysian Food Regulations, 1985
^^ World Health Organisation Recommended Guidelines, 2007

There was a significant positive ($p < 0.01$) relationship between concentrations of copper and zinc in cabbage with concentrations of these two elements in soil ($r = 0.9567$ and $r = 0.8177$), respectively (Fig. I). Correlation was not computed for lead or cadmium as cadmium was not detected in all cabbage samples and lead only detected in a handful of cabbage samples.

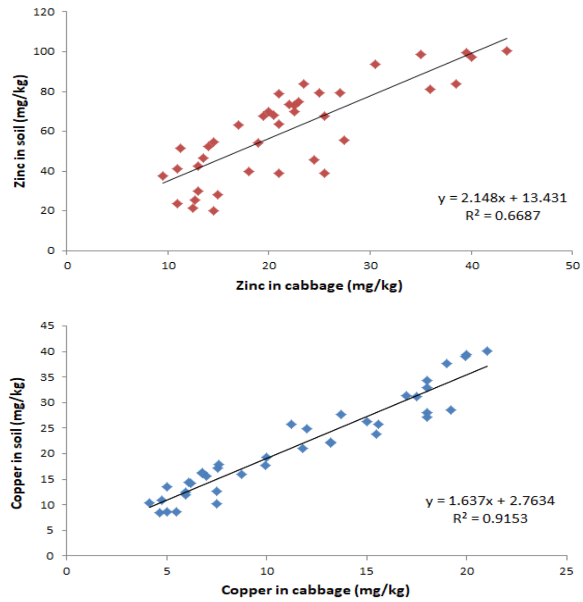

Fig. I: Correlation of copper and zinc concentration in soil and cabbage. The dot plot analysis shows a positive correlation between copper and zinc concentrations in soil and cabbage.

Dietary Exposure and Potential Health Risks

Using the suggested average consumption amount of cabbage for Malaysians the Chronic Daily Intake (CDI) of metals was calculated. The CDI of the measured metals from consumption of cabbage from new and old farms are presented in Table 3. The exposure of metals through the consumption of cabbage was in the ascending trend of lead < copper < zinc. CDI for cadmium was not calculated as cadmium concentrations were below detection limits in all cabbage samples. The Tolerable Daily Intake (TDI) is an estimated quantity of a substance which a person may be exposed to without an expectation of a health risk. The contribution of metals through the average serving sizes of cabbage are well within the suggested TDI values (22) and are shown in Table 3. Individual Target Hazard Quotients for the three measured metals from the consumption of cabbage are shown in Fig. 2. There is no calculated potential health risk to consumers from ingestion of cabbage from both new and old farms as the THQ values for all metals were lower than one.

Table 3: Chronic daily intakes (mg/kg BW/day) of metals by consuming cabbage

Metal	Tolerable Daily Intake (TDI) (ug/kg BW/day)	New Farm		Old Farm	
		Daily Intake (ug/kg BW/day)	% of TDI	Daily Intake (ug/kg BW/day)	% of TDI
Copper	500	0.0736	0.02	24.25	4.85
Lead	3.57	0.0039	0.11	0.0613	1.71
Zinc	300-1000	24.43	2.44 - 8.14	42.59	4.26 - 14.19

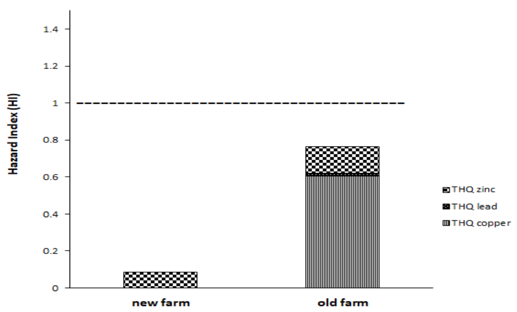

Fig. II: Hazard Index via cabbage consumption from old and new farms. The community may experience a potential accumulative risk if the HI is either equal to or bigger than 1.

DISCUSSION

Large quantities of mineral fertilizers are routinely used during the farming process to deliver adequate amounts of nitrogen, phosphorus, and potassium (NPK) for crop development. These fertilizers especially those of uncertain quality frequently contain trace quantities of metals as impurities, contaminating and increasing the metal content in soils after regular application (23). Metals that have no known biological function, such as cadmium and lead, may pose potential health risks to humans or other living organisms in the ecosystem thru either contact with contaminated soil or ingestion of bio-accumulated metals in the food chain. The non-health risks of soil contamination include contaminated agricultural areas causing food safety issues, problems in marketability of products and land usability for agricultural production leading to food insecurity (24).

Overall, this study shows that there was accumulation of metals at the Monteki agricultural area in both soil and cabbage in the order of zinc > copper > lead > cadmium. High accumulation of zinc and copper in soil is because of NPK fertilizer application (25). In this study area, workers were using NPK fertilizer labelled as “Cap Dacing”. One of the most important factors to be considered is the effect of long term application of fertilizer without any treatment. The longer the application of fertilizer, there will be more accumulation of metals in soil (6). In addition, soils with pH lower than 7 will accumulate metals (23). Additionally continuous use of fertilizer can contribute to soil acidification (26). Hence, repeated use of fertilizer in the study site has caused a decrease in soil pH leading to a higher accumulation of metals in old farms compared to new farms.

The accumulation of metals in vegetables is related to the transfer rate and net absorption of the metals from soil. Among leafy vegetables, fruit vegetables and root vegetables, accumulation of metals is much higher in leafy vegetables compared to the other two types of vegetable (8) due to the translocation rate and transpiration rate (9). Cabbage samples in this study had the capacity to absorb metals accumulated in the soil and transfer those metals to the edible part of the cabbage. Levels of metal in cabbage increased as a result of increasing concentration in soil. The uptake of lead was the lowest for cabbage from both locations, whereas cadmium was not detected in all cabbage samples. This may be attributed to lower levels of these two metals in the top soil sampled, presence of soil organic matter and soil conductivity. The presence of metals in edible vegetables is not all damaging as elements such as copper and zinc are critical for the maintenance of normal human body functions. However, chronic exposure to excess zinc is suspected to affect cardiac function whereas liver damage may be triggered by chronic copper exposure. The cabbage samples from the study location are found to be suitable sources of dietary copper and zinc, albeit within the permissible levels. Nevertheless, it is important to make sure that the amount of metals accumulated in cabbage is monitored to ensure low possibility of transfer through the food chain.

Cabbage is reported to be the third largest crop grown in Malaysia (1) with an average weekly consumption rate in both Peninsular Malaysia and East Malaysia (14). Consequently, to determine whether the amount of metals accumulated in cabbage from this study area can cause adverse health effects to the population, the chronic daily intake (CDI) and hazard index (HI) was calculated to determine the combination risk from copper, lead and zinc exposure. A normal consumption of cabbage from this location does not contribute significant amounts of copper, lead or zinc as the chronic daily intake of metals was less than 15% of the Tolerable Daily Intake. As the Hazard Index is less than 1 for both locations it is currently still safe to consume cabbage from this location. It should be noted that continuous application of fertilizer into the soil for long periods of time can increase the accumulation of metals in the soil. It is not impossible that in the future there will be an accumulation of cadmium or other trace metals deposited in cabbage exceeding the standard limits if there is no precautionary step or treatment done to the soil. An increase the organic matter content and soil conductivity has been known to be associated with immobilization of heavy metals for plant uptake (27-29). Thus, if there were no improvement on the technique of using the fertilizer and no treatment been applied, the accumulation in soil will exceed the standards and may affect metal concentration in vegetables as well.
Moreover it should be noted that consumers are exposed to metals from consuming various other vegetables and food such as fish and fruit. The presence and bioaccumulation of metals in aquatic organisms such as shrimp, fish and shellfish (30) and fruits (31) have also been presented. Subsequently, consumers may also be exposed to metals via dust inhalation or dermal contact (32). Although, the HI was below the cut off value, there are various other sources of metals that consumers might be accumulatively exposed to daily that may affect their health.

CONCLUSION

The results of this study revealed varying concentrations of cadmium, copper, lead and zinc in the cabbage and soil obtained from the Monteki agricultural area of Sabah, Malaysia. There was a positive correlation between metal content in the cabbage and soil for copper and zinc which suggest common source of inputs. Although, the present levels of the metals were lower than their regulatory limits in vegetable, continuous accumulation is a concern for produce safety and quality. Though levels of metals increased significantly in old farms, they were still below acceptable limits. These findings verify that the longer the application of fertilizer in soils the higher the accumulation of metals which can directly influence its accumulation in cabbage.

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