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Elemental variations and safety assessment of commercial onions (*Allium cepa*) by inductively coupled plasma-mass spectrometry and chemometrics

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Abstract

The concentrations of Cd, As, Pb, Se, Fe, Mn, Zn, and Cu in acid-digested onion samples were determined by inductively coupled plasma mass spectrometry, and the corresponding elemental profiles were evaluated by using chemometric techniques. Results from principal component analysis and hierarchical cluster analysis revealed that the spatial variations were highly associated with As, Se, Pb, Zn, and Cd, which advocated dominance of the site related impacts over colour variant of the onions. Such variation pattern could be partly attributed to the consequences of industrialisation and/or urbanisation. From the safety assessment data, concentrations of hazardous elements such as As, Cd, and Pb, were found beyond the World Health Organisation and Food and Agricultural Organisation permissible levels, but within the Malaysian regulatory limits except for Pb. The presence of elevated levels of toxic elements beyond the reference limits in the onion varieties could be of health concern if ingested.

Introduction

Allium is recognised as one of the largest monocotyledon genera that comprises of more than 600 species, and several species are cultivated for various purposes (Li *et al.*, 2010; Choi and Oh, 2011). Among them, *Allium cepa* (bulb onions) is believed to offer health and nutritional benefits. Nowadays, onions have great economic importance with an annual global production of about 76 million tonnes (Lanzotti, 2006). The global trend of onion production increases over years. India and China used to be the leading producing countries which together contributed approximately half of the world supply (Silva Dias, 2011). Therefore, it is not surprising that the raw onions available in the Malaysian market mainly originated from the above countries, while some are imported from Australia, New Zealand, and the Netherlands (Currah, 2002). From the consumer's viewpoint, it is believed that quality and/or the tagged price of the produce mainly depend on the country of origin. In fact, exploring the relations between characteristics of onions and their origins is a complex process.

Commercially grown onions are generally available in four main colour varieties, i.e. red, yellow, brown, and white (Kim *et al.*, 2004). The two former varieties can be easily found in the Malaysian market because they serve diverse culinary purposes based upon their inherent features (Farr *et al.*, 2015). To a large extent, the corresponding sensory properties are attributed to chemical variability that is in turn associated with the cultivars, and additional deviations are derived from the geographical origins (Havey *et al.*, 2002). In this regard, environmental and agronomic conditions are the main factors that affect the botanical sources (Rodríguez Galdón *et al.*, 2009). Such variation suggests potential distinction of onions properties/quality on a regional basis (Yang *et al.*, 2004). This could be the reason for the increasing consumer preference on produce that originate from specific regions and are willing to pay more for that on top of food safety concerns (Shaheen *et al.*, 2016). Among the chemical variables, the

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elemental fingerprint has gained much attention from researchers as it provides a reasonable reflection on corresponding environment, climate, soil nature, fertiliser, and degree of plant maturity at the time of harvest (Lake *et al.*, 1984; Yusuf *et al.*, 2003).

Besides as geographical indicator, trace elements are known to play vital roles in food safety and human health (Divrikli *et al.*, 2003; Sobukola *et al.*, 2010). Trace elements can be placed into three distinct categories, namely essential (Se, Co, Cu, Mn, Fe); semi essential (V and Ni) and toxic elements (As, Hg, Cd, Pb) (Muñoz-Olivas and Cámara, 2001). The essential and semi essential elements could well produce toxic effect upon excessive intake (Çelik and Oehlenschläger, 2007). Due to their non-biodegradability, toxic elements may pose a serious threat to humans as well as animals when accumulated in different body parts (D'Mello, 2003; Jafarian-Dehkordi and Alehashem, 2013). Similarly, the exposure to such hazardous elements via onion consumption has raised global concerns. To safeguard the health of the citizens, the food production and consumption regulatory agencies in Malaysia adhere to the limits set by the World Health Organisation and Food and Agricultural Organisation (WHO and FAO, 2004), and Malaysia Food Act 1983 and Food Regulations 1985 (Malaysia, 1994).

The objective of the present work is not limited to safety assessment of the concentrations of As, Cd, Pb, Mn, Se, Fe, Cu, and Zn in the commercially available onion samples from the local market with reference to their regulatory limits, but also to explore the corresponding variation patterns by chemometric techniques i.e. principal component analysis (PCA) and hierarchical cluster analysis (HCA). These techniques have been previously demonstrated to be useful in food authentication and quality control (Osorio *et al.*, 2013; Moghadam *et al.*, 2016; Zain *et al.*, 2016; Ikram *et al.*, 2019).

Materials and methods

Sampling

In 2017, onion samples originated from China, India, Australia, New Zealand, and Holland were purchased from a supermarket in Kuala Lumpur. The samples were kept inside polythene bags and transported to the laboratory immediately after purchase.

Sample preparation

The sample bulbs were washed several times with ultrapure water (UPW), sliced into a thickness of ~4 mm, and oven-dried at 100°C for 12 h to a consistent weight. The dried samples were ground into fine particles and stored in an amber bottle under a desiccator.

Quality assurance

All chemical reagents were of analytical grade or better quality. Suprapur® 65% HNO_3 and 70% H_2SO_4 solutions were purchased from Merck (Germany). Multi-elemental calibration standard solution of 1000 mg/L for Fe, and 10 mg/L for As, Cd, Pb, Mn, Se, Cu, and Zn in 5% nitric acid were purchased from Agilent Technologies (Newcastle). Ultrapure water from PURELAB® UHQ II system (ELGA®, UK) was used for reagent preparation throughout the study. Quality control samples were analysed after every eight samples to confirm and demonstrate the validity of the previous runs. All the plastic-wares used were soaked overnight in 10% HNO_3 (v/v), and rinsed twice (Low *et al.*, 2011).

Acid digestion

Approximately 1 g of the powdered sample was treated with a mixture of 10 mL of 65% HNO_3 and 5 mL of 70% H_2SO_4. Briefly, the digestion was carried out at ~80°C until a transparent solution was obtained. After cooling, the digestate was filtered into a volumetric flask for appropriate dilution before being subjected to elemental analysis.

Elemental analysis

The concentrations of As, Pb, Mn, Zn, Fe, Se, Cu, and Cd were determined simultaneously by Agilent Technologies inductively coupled plasma-mass spectrometer (ICP-MS 7500Ce) in triplicates. The instrument was tuned and calibrated by using freshly prepared standard solutions, whereby the corresponding standard and sample solutions were introduced via an auto sampler (ASX 500). In order to maintain the analytical performance of the instrument, the operating conditions were monitored and tuned following the practice described by Low *et al.* (2012).

Data analysis

The ICP-MS data matrix (108 × 8) which corresponded to 108 entries by eight elements was subjected to statistical analyses. The descriptive statistics were calculated using MSExcel 2013, and multivariate analyses including PCA and HCA were carried out with JMP Pro 12. PCA and HCA were employed to reveal the underlying structure and natural grouping based on the elemental characteristics of the samples (Low *et al.*, 2016; Koki *et al.*, 2017).

Table 1. Mean concentrations of metals in onion samples, µg/g (dry weight basis).

Variety	Origin	As	Cd	Pb	Mn	Se	Fe	Cu	Zn	Source
Red	Australia	5 ± 4(0.72 - 11.69)	2 ± 1(2.11 - 2.24)	29 ± 4(24.32 - 33.29)	7 ± 2(5.17 - 8.78)	7 ± 1(5.47 - 9.37)	42 ± 3(38.91 - 45.28)	12 ± 3(9.85 - 17.28)	17 ± 4(12.75 - 22.35)	The present work
	China	60 ± 3(54.87 - 62.13)	3 ± 1(2.02 - 5.13)	38 ± 7(27.57 - 49.72)	6 ± 1(4.30 - 7.22)	8 ± 2(5.87 - 11.05)	41 ± 7(32.67 - 48.60)	10 ± 1(8.76 - 11.64)	34 ± 3(29.96 - 36.50)	
	India	5 ± 3(1.59 - 8.67)	3 ± 1(2.06 - 4.73)	35 ± 3(29.80 - 38.32)	5 ± 1(3.66 - 7.13)	6 ± 1(6.23 - 6.60)	45 ± 2(41.31 - 47.86)	10 ± 1(9.29 - 11.24)	15 ± 3(12.71 - 21.05)	
Yellow	Australia	9 ± 3(4.22 - 13.18)	2 ± 1(2.21 - 2.63)	29 ± 3(24.57 - 32.80)	5 ± 2(2.41 - 8.59)	6 ± 3(5.71 - 6.58)	39 ± 4(32.41 - 43.62)	10 ± 2(8.39 - 14.17)	27 ± 2(23.10 - 29.58)	
	Holland	3 ± 1(2.59 - 4.29)	2 ± 1(2.24 - 2.52)	34 ± 2(32.71 - 36.57)	6 ± 2(3.86 - 9.49)	6 ± 3(5.90 - 6.78)	40 ± 2(36.42 - 43.67)	10 ± 1(9.39 - 15.79)	16 ± 1(11.46 - 21.92)	
	New Zealand	7 ± 3(0.10 - 10.25)	2 ± 1(2.10 - 2.37)	30 ± 4(34.56 - 26.15)	4 ± 2(1.72 - 6.51)	6 ± 3(2.25 - 6.66)	43 ± 5(33.86 - 47.26)	10 ± 1(9.16 - 11.03)	16 ± 1(13.74 - 18.04)	
Unspecified	Argentina	92 ± 7	-	-	-	-	363 ± 206	17 ± 3	207 ± 59	Pintera et al. (2018)
		73 ± 4	-	-	-	-	418 ± 204	10 ± 1	7 ± 3	
	Croatia	-	0.30 ± 0.10	0.17 ± 0.01	3 ± 1	-	-	9 ± 4	42 ± 9	Stančić et al. (2016)
	Ethiopia	-	0.60 ± 0.10	< 0.5	9 ± 1	-	-	3.20 ± 0.10	18 ± 1	Alloway (1990)
		-	0.20 ± 0.01	4.90 ± 0.80	5.40 ± 0.10	-	28 ± 1	2.50 ± 0.10	3 ± 1	Gebrekidan et al. (2013)
	Nigeria	-	0.22 - 0.89	2.00 - 9.50	1.00 - 6.60	-	-	0.34 - 1.00	2.00 - 4.55	Abdullahi et al. (2008), Akan et al. (2009)
	Pakistan	-	-	-	55 ± 34	-	-	5 ± 1	14 ± 8	Rehman et al. (2018)
	Saudi Arabia	-	0.90 ± 0.10	3.50 ± 0.10	8 ± 1	-	50 ± 3	3.30 ± 0.10	20 ± 1	Ali and Al-Qahtani (2012)
		-	1.00 ± 0.10	4.20 ± 0.10	19 ± 1	-	52 ± 1	10.00 ± 0.30	22 ± 1	
		-	0.90 ± 0.10	4.40 ± 0.10	9 ± 1	-	32 ± 1	4.9 ± 0.2	11 ± 1	
		-	1.10 ± 0.10	3.10 ± 0.20	10 ± 1	-	67 ± 2	7.3 ± 0.1	23 ± 1	
	Turkey	-	0.97 ± 0.01	8.70 ± 0.10	-	-	-	54 ± 2	21 ± 4	Demirezen and Aksoy (2006)

- : Not reported.

Table 2. Mean concentrations of metals in onion samples, µg/g (fresh weight basis).

Variety	Origin	As	Cd	Pb	Mn	Se	Fe	Cu	Zn
Red	Australia	0.5 ± 0.4(0.07 - 1.10)	0.2 ± 0.1(0.10 - 0.24)	2.9 ± 0.4(2.28 - 3.27)	0.7 ± 0.2(0.53 - 0.90)	0.7 ± 0.2(0.51 - 0.64)	4.2 ± 0.4(3.73 - 4.75)	1.2 ± 0.3(0.96 - 1.62)	1.7 ± 0.5(1.19 - 2.23)
	China	4 ± 1(3.86 - 5.38)	0.2 ± 0.1(0.15 - 0.33)	2.6 ± 0.4(2.34 - 3.53)	0.5 ± 0.1(0.38 - 0.53)	0.6 ± 0.1(0.38 - 0.72)	2.9 ± 0.2(2.62 - 3.18)	0.7 ± 0.1(0.57 - 0.83)	2.5 ± 0.3(2.23 - 2.91)
	India	0.6 ± 0.4(0.19 - 1.07)	0.4 ± 0.2(0.24 - 0.61)	4.3 ± 0.4(3.69 - 4.95)	0.7 ± 0.2(0.43 - 0.88)	0.8 ± 0.1(0.74 - 0.83)	5.6 ± 0.4(4.90 - 5.93)	1.2 ± 0.1(1.20 - 1.39)	1.9 ± 0.4(1.57 - 2.60)
Yellow	Australia	0.9 ± 0.3(0.41 - 1.29)	0.2 ± 0.1(0.22 - 0.26)	3.0 ± 0.4(2.41 - 3.56)	0.6 ± 0.3(0.26 - 0.91)	0.6 ± 0.1(0.60 - 0.64)	4.1 ± 0.4(3.51 - 4.73)	1.1 ± 0.2(0.91 - 1.51)	2.9 ± 0.3(2.35 - 3.18)
	Holland	0.3 ± 0.1(0.25 - 0.92)	0.2 ± 0.1(0.19 - 0.24)	3.1 ± 0.3(2.77 - 3.60)	0.5 ± 0.2(0.38 - 0.80)	0.6 ± 0.1(0.50 - 0.67)	3.6 ± 0.2(3.36 - 3.92)	1.0 ± 0.2(0.86 - 1.34)	1.3 ± 0.3(0.97 - 1.86)
	New Zealand	0.7 ± 0.3(0.10 - 0.95)	0.2 ± 0.1(0.20 - 0.25)	3.0 ± 0.5(2.44 - 3.42)	0.4 ± 0.2(0.16 - 0.61)	0.6 ± 0.1(0.54 - 0.66)	4.2 ± 0.5(3.50 - 4.75)	0.9 ± 0.1(0.74 - 1.04)	1.6 ± 0.2(1.28 - 1.87)
WHO and FAO (2004)		0.1	0.05	1.5	-	-	-	30	100
Malaysia Food Act 1983 and Food Regulations 1985 (Malaysia, 1994)		1.0	1.0	2.0	-	-	-	10	100

Elemental concentrations are reported at 95% confidence intervals.

- : Not reported.

Results and discussion

Safety assessment

Metal elements enter human body mainly via the food web. However, their accumulation in excessive levels may pose undesired health risks. In the present work, the comparisons with permissible limits would serve as a preliminary basis for safety assessments. Table 1 reports the total elemental concentrations of the onion samples measured on dry weight basis together with some findings from similar studies. Meanwhile Table 2 displays the concentrations based on fresh weight and their reference limits.

Based on measurement results, total As in the onion samples ranged from 3 to 60 µg/g. The highest concentration of (60 ± 3) µg/g was observed in the red onions imported from China, which shared the same order of magnitude as those collected from the onion fields in Jáchal, Argentina (Pintera *et al.*, 2018). Based on the fresh weight concentrations, all As contents were found beyond the WHO and FAO (2004) recommended value. However, they were still below the limit established in the Malaysia Food Act 1983 and Food Regulations 1985 (Malaysia, 1994), except the abovementioned sample from China. Such elevated level of As in the crop suggested the major attribution from the field site on which they were grown, to some extent, urbanisation and/or industrialisation across the production region (Yuan *et al.*, 2014).

The concentrations of Cd in the onion samples were found between 2 µg/g and 3 µg/g where no statistical evidence in difference was triggered by the colour variants. Although the mean concentrations found were generally higher than literature values that were reported at sub-ppm levels, the fresh weight concentrations still fell within the regulation limit. There are many potential sources of Cd, apart from the direct inputs from agricultural soils, irrigation water and fertilisers; the accumulation is also indirectly linked with atmospheric deposition, leaching, and improper waste disposal from related industries (Alloway *et al.*, 1990; Zhang *et al.*, 1998; Norton *et al.*, 2015; Rizwan *et al.*, 2017).

High concentrations of Pb were detected in the onion samples (29 to 38 µg/g). The levels found were much greater than other reported values and even surpassed the maximum permissible levels listed in Table 1. This thus poses potential health risk associated with chronic Pb exposure via onion consumption. Based on literature reports, it is believed that the high levels of Pb are due to contaminations from automobile and industrial activities (Nabulo *et al.*, 2006; Stančić *et al.*, 2016; Orisakwe *et al.*, 2017).

The concentrations of Cu and Zn were 10 to 12 µg/g and 15 to 34 µg/g on dry weight basis,

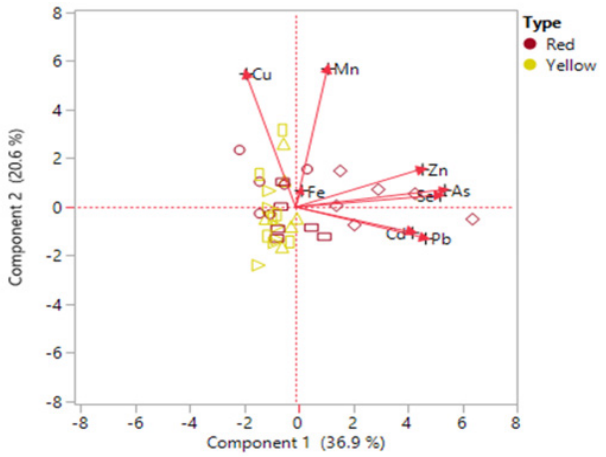

Figure 1. PCA biplot of onion samples.

respectively, where the red onions from China demonstrated the highest Zn content. On fresh weight basis, their concentrations were well below the permissible levels set by WHO and FAO (2004), Malaysia Food Act 1983 and Food Regulations 1985 (Malaysia, 1994). Mn, Se and Fe are essential trace elements that are not usually of regulatory concern. However, these elements might be harmful when excessively consumed; particularly Se whose toxicity depends on its chemical species, whereby inorganic forms are more toxic than the organic forms (Mertz, 1981; Goldhaber, 2003). The concentrations of Mn and Fe in the onion samples were 4 to 7 µg/g and 39 to 45 µg/g, respectively, which were consistent with the common ranges reported elsewhere (Mohamed *et al.*, 2003; Ali and Al-Qahtani, 2012; Gebrekidan *et al.*, 2013). Total Se ranged between 6 and 8 µg/g, indicating that onion is one of the significant sources of Se. In addition, the association between Fe content and colour variant was not observed in the present work.

Exploratory of elemental variation

To interpret variances among the samples and elemental variables, PCA transformed the ICP-MS dataset into a set of uncorrelated principal components (Zhao *et al.*, 2012). The eigenvalues of the eight principal components (PCs) were obtained, in which PC1 and PC2 together explained about 57.5% of the total variation in the entire data set. From Figure 1, it was observed that several red onion samples (from China) and elemental variables were dragged towards the area with positive loading on PC1. Such dispersion pattern was mainly associated with the deviations in the concentrations of As, Cd, Pb, Se, and Zn, which disclosed that biological factor, i.e. inherent colour, was not a key controlling factor for the elemental variability in the onions. This partly explains the overlapping of the remaining red onion varieties that

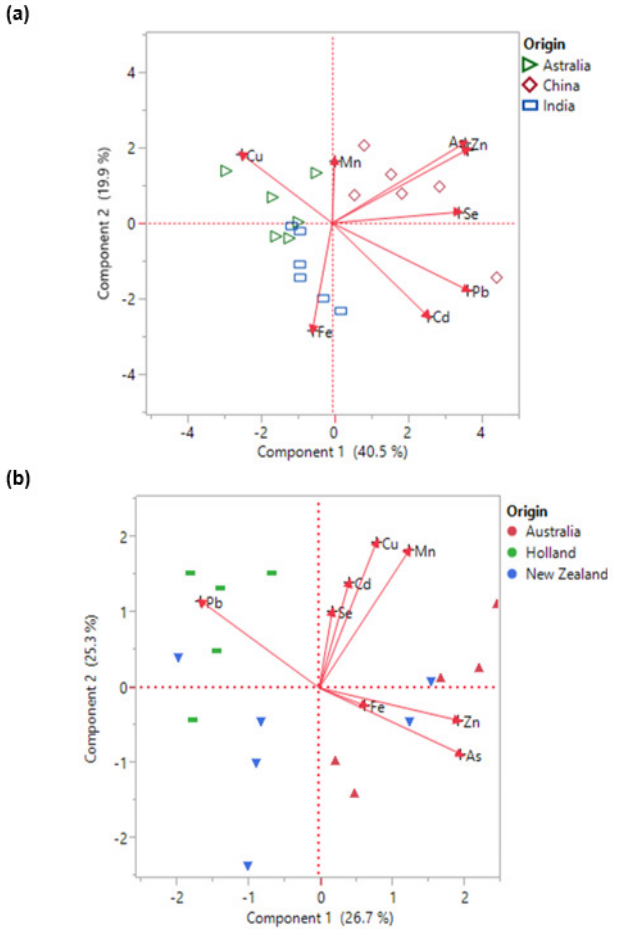

Figure 2. (a) biplot of red onion samples, and (b) biplot of yellow onion samples.

were mapped together with the dominance of yellow onion variety on the opposite side. Based on the scores and loading patterns, the clustering observed was more likely attributed to the site related impacts on bioavailability of metal elements and their uptake rates (Patra *et al.*, 2004; Aris *et al.*, 2013). Similar findings were reported on different varieties of rice, berries, and vegetables which linked the elemental variations with soil nature, agricultural practices and environmental pollution (Meharg *et al.*, 2008).

By blocking the nuisance factor, i.e. variety of colour, Figure 2(a) illustrates the elemental variability in which PC1 (40.5%) and PC2 (19.9%) together explained about 60.4% of the total variation among the red onion samples. Again, it shows that As, Cd, Pb, Se, and Zn with strong positive loadings on PC1 were noticeably associated with the samples imported from China. The finding was consistent with the results listed in Table 1, which put forward the potential elemental inputs from the growing industrial activities in the region; in addition to the underlying geographical factors (Plessi *et al.*, 2007). The biplot also indicates the potential association between the onion samples from India with Fe and those from Australia with Cu. Considering the yellow

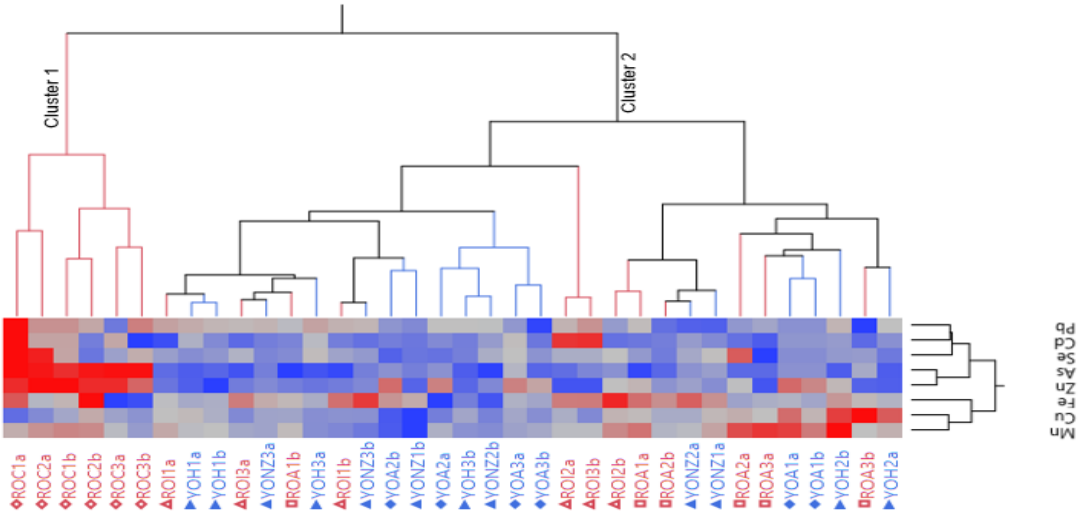

Figure 3. Two-way clustering of onion samples.

onion samples, Figure 2(b) shows strong positive loadings of As and Zn, and negative loading of Pb on PC1, while PC2 with high loadings of Cu and Mn. These notable differences again expressed the site-specific influences besides the natural variations. In this regard, elemental variability could serve as a basis for traceability and discrimination between the origin of onion produce (Conti *et al.*, 2000; Fernández-Cáceres *et al.*, 2001; Berna *et al.*, 2009; Zain *et al.*, 2016).

Figure 3 is derived from two-way HCA results which depict the clustering patterns of both samples and elemental variables. Two heterogeneous distinct sample clusters were observed, where the red onion samples from China were well partitioned from the rest owing to their As, Zn, Cd, Se, and Pb contents, as outlined in the heat map and secondary dendrogram. The outcomes were consistent with the trends revealed in previous PCA findings that supported the dominance of site related impacts which are likely to be attributed to the massive industrialisation and/or urbanisation (Wong *et al.*, 2002; Li and Jia, 2018).

Conclusion

The present work provides the background concentrations of selected elements in commercial onion samples from the Malaysian market. The concentrations of hazardous elements namely As, Cd, and Pb in the samples were found beyond the recommended values by WHO and FAO (2004), however, they were within the limits set in the Malaysia Food Act Regulation 1983 and Regulations 1985 (Malaysia, 1994), except for Pb. Thus, consumers may be at potential risk of Pb exposure. The employed pattern recognition techniques provided an objective interpretation of the ICP-MS results by exploring the relationships between elemental variability and onion samples. The clustering patterns by both PCA and HCA revealed the dominance of site related impacts over the inherent biological variations on the elemental fingerprints, whereby the elemental profile of onion samples from China seemed to be associated with high degree of industrial activities.

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