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Heavy metal exposure from cooked rice grain ingestion and its potential health risks to humans from total and bioavailable forms analysis

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ABSTRACT

Heavy metal in rice studies has attracted a greater concern worldwide. However, there have been limited studies on marketed rice samples although it represents a vital ingestion portion for a real estimation of human health risk. This study was aimed to determine both total and bioaccessible of trace elements and heavy metals (Cd, Cr, Cu, Co, Al, Zn, As, Pb and Fe) in 22 varieties of cooked rice using an inductively coupled plasma-optical emission spectroscopy. Both total and bioaccessible of trace elements and heavy metals were digested using closed-nitric acid digestion and Rijksinstituut voor Volksgezondheid en Milieu (RIVM) *in vitro* digestion model, respectively. Human health risks via Health Risk Assessment (HRA) were conducted to understand exposure risks involving adults and children representing Malaysian population. Zinc was the highest while As was the lowest contents for total and in their bioavailable forms. Four clusters were identified: (1) Pb, As, Co, Cd and Cr; (2) Cu and Al; (3) Fe and (4) Zn. For HRA, there was no any risks found from single element exposure. While potential carcinogenic health risks present for both adult and children from single As exposure (Life time Cancer Risk, LCR > 1×10^{-4}). Total Hazard Quotient values for adult and children were 27.0 and 18.0, respectively while total LCR values for adult and children were 0.0049 and 0.0032, respectively.

1. Introduction

Humans are exposed to trace element and heavy metal through the daily ingestion of food and beverages. Trace element and heavy metal ingestion through rice is a crucial to be investigated as rice has been dominating staple food for more than two billion people especially from Asia (World Health Organization., 2003). World Health Organization (2003) has indicated that average daily intake of rice per person ranges from 9 g in west countries and 600 g in Asia which clearly showed that rice contribution to toxic elements exposure in human is crucial to be investigated. International Agency for Research on Cancer (2012) has classified arsenic (As), cadmium (Cd) and lead (Pb) are both carcinogenic and non-carcinogenic to humans while chromium (Cr), cobalt (Co), copper (Cu), iron (Fe), aluminium (Al) and zinc (Zn) are non carcinogenic to humans. Retention of Cd in human kidney can cause kidney failure while As can cause severe health effects such as malignant neoplasms, malnosis and depigmentation (Abedin, Cresser, & Meharg, 2002).

Toxic elements in rice studies have attracted a great concern worldwide involving paddy field and marketed rice samples. Rice has ability to absorb toxic element from soil and water in paddy field more effectively than other crops. This is due to paddy plants is grown in water flooded conditions which permit toxic elements to be taken up by its root and accumulated in rice (Huang et al., 2016). Anaerobic condition in paddy field soil has resulted in higher mobilization rates and increase bioaccessible form of toxic elements in rice. Since toxic elements have the ability to accumulate and non-biodegrable, the presence of toxic element concentrations in marketed rice is largely unavoidable (Morekian, Mirlohi, Azadbakht, & Maracy, 2013). With increasing trend of annual rice consumption indicated by OECD/Food (2015), understanding toxic element exposure through marketed rice will give a clear view of exposure involving population and its exposure risks to human health. In Malaysia, most of the rice studies were conducted involving field rice samples related to pesticides, insecticide, fertilizers and organic contamination. Although study done by Salim, Elias, and Wood (2010) has focused on multiple toxic elements in rice varieties yet limited information was available on human health in the context of population and consumers exposure risks. So far, study done by Omar, Praveena, Aris, and Hashim (2015) involving cooked rice has applied human health risk assessment with an inclusion of bioaccessible form of trace element and heavy

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metal for realistic health risk estimation in Malaysia. However, study on both total and bioaccessible form of trace element and heavy metal in rice is still unclear to understand the similarities and behaviors in chemical properties of trace element and heavy metal in human gastrointestinal tract which will lead to human health risks (non-carcinogenic and carcinogenic) in long term accumulation.

According to Versantvoort, Oomen, and Kamp (2005), total trace element and heavy metal concentrations in rice overestimates the potential human health risks with a gap in understanding the actual human health risks as well as inadequate for toxic effect understanding. Bioaccessible form of trace element and heavy metal is defined as the fraction of total trace element and heavy metal concentrations present in a specific environmental compartment within a time and being uptaken by organisms or plants from direct environment via ingestion (Peijnenburg & Jager, 2003). Determination of bioaccessible form of trace element and heavy metal can be done via *in vitro* digestion models (Yuswir, Praveena, Aris, Ismail, & Hashim, 2015). In rice studies, bioaccessible form of trace element and heavy metal can be determined by Rijksinstituut voor Volksgezondheid en Milieu (RIVM) *in vitro* digestion model as since it mimics quite close to the physiological condition in human body. The model involves three compartments that were oral cavity, stomach and small intestine, with parameters (pH, residence time and particle size) based on human physiologically condition (Omar et al., 2015; Versantvoort et al., 2005). Thus, both total and bioaccessible form of trace element and heavy metal concentrations are crucial to understand the accumulation and human health risks via rice ingestion.

Therefore, the main objective of this study was to determine both total and bioaccessible trace element and heavy metal concentrations in varieties of cooked rice samples using ICP-OES. This study also classifies the similarities and behaviors in chemical properties of trace element and heavy metal by using Hierarchical Cluster Analysis (HCA). Carcinogenic and non carcinogenic health risks exposure to Malaysian population (adult and children) via ingestion pathway were also determined by using health risk assessment. This study output opens to an accurate estimation on human health risk in varieties of cooked rice using *in vitro* digestion model with clear understanding on trace element and heavy metal contamination level in rice and exposure risks to Malaysian population.

2. Materials and methods

2.1. Sampling and analysis

Convenience sampling was chosen as a sampling method in this study due to easy accessibility, and availability at a given time (Etikan, Musa, & Alkassim, 2016). A total of 22 rice varieties were randomly purchased from local consumer products' superstores in Malaysia based on the most and least rice varieties preferred by Malaysian (Musa, Othman, & Fatah, 2011). The origin of rice samples was taken from the package's label as given in Table 1. All rice samples were cooked based on method by Salim et al. (2010). A total of 50 g of raw rice sample from each rice variety sample was weighed and washed three times with deionised water. Then, the rice sample was cooked with 100 mL deionised water (1:2 ratio) for white rice, and with 150 mL deionised water (1:3) for other rice variety samples. The rice samples were allowed to cook until there was no water left. As the rice samples were cooked, all the rice samples were oven dried at 65 °C for 48 h in order to obtain the dry matter content. The rice samples were then grounded manually with pestle and mortar to avoid cross-contamination and on a cleanliness standpoint. Then, the rice samples were allowed to pass a 0.25 mm mesh sieve (No 60 mesh sieve). Lastly, the milled rice samples were kept in sealed bags at 4 °C until the digestion process took place.

Table 1
Cooked rice samples.

	Rice varieties	Origin
Most preferred	Local unpolished brown rice	Kedah, Malaysia
	Local fragrant rice	Selangor, Malaysia
	Siam fragrant rice	Thailand
	Thai fragrant rice	Thailand
	Basmathi rice	Pakistan
	Parboiled rice	Selangor, Malaysia
	Local rice (5% broken)	Selangor, Malaysia
	Local white rice	Kedah, Malaysia
	Imported white rice	Thailand
	Special glutinous rice	Selangor, Malaysia
	Milky glutinous rice	Kedah, Malaysia
	Thai glutinous rice	Thailand
Least preferred	Imported brown rice	Thailand
	Red rice	Thailand
	Mix grain rice	India
	Ponni rice	India
	Hill rice	Perak, Malaysia
	Siam rice	Thailand
	Organic black rice	Taiwan
	Special Siam rice	Thailand
	Calrose grain rice	Japan
	Steamed rice	India

2.2. Total and bioaccessible of toxic element analysis

Total trace element and heavy metal concentrations were analyzed using the closed-acid digestion method using a Parr Acid Digestion Vessel to avoid loss or contamination of the analyte. A total of 0.4 g of rice sample was accurately weighed into a Parr Acid Digestion Vessel, followed by addition of 20 mL of nitric acid (ACS reagent, >90%), and then heated from 25 °C to 95 °C for 45 min and maintained at 95 °C for 90 min using heating plate. After complete digestion, the sample mixture was filtered through Whatman 0.45 μm filter paper.

To analyze the bioaccessible of trace element and heavy metal, the methods applied by Versantvoort et al. (2005) and Yang, Zhang, and Li (2012) were adopted. The RIVM model includes simulated digestive processes starting from the mouth, stomach, and finally to the small intestine. The chemicals and reagents used for artificial saliva included 10 mL of potassium chloride (KCl), 10 mL of potassium thiocyanate (KSCN), 10 mL of sodium dihydrogen phosphate (NaH_2PO_4), 10 mL of disodium sulfate (Na_2SO_4), 1.7 mL of sodium chloride (NaCl), 20 mL of sodium bicarbonate ($NaHCO_3$), 8 mL of urea, 290 mg of amylase, 15 mg of uric acid, and 25 mg of mucin. Gastric juice contained 15.7 mL of NaCl, 3.0 mL of NaH_2PO_4, 9.2 mL of potassium chloride (KCl), 18 mL of calcium chloride dehydrate ($CaCl_2{\cdot}2H_2O$), 10 mL of ammonium chloride (NH_4Cl), 6.5 mL of hydrochloric acid (HCl), 10 mL of glucose, 10 mL of glucuronic acid, 3.4 mL of urea, 10 mL of glucosamine hydrochloride, 1 g of bovine serum albumin (BSA), 2.5 g of pepsin, and 3 g of mucin. Duodenal juice contained 40 mL of NaCl, 40 mL of $NaHCO_3$, 10 mL of potassium dihydrogen phosphate (KH_2PO_4), 6.3 mL of KCl, 10 mL of magnesium chloride ($MgCl_2$), 180 μL of HCl, 4 mL of urea, 9 mL of calcium chloride dehydrate ($CaCl_2{\cdot}2H_2O$), 1 g of BSA, 9 g of pancreatin, and 1.5 g of lipase. Bile juices contained 30 mL of NaCl, 68.3 mL of $NaHCO_3$, 4.2 mL of potassium chloride (KCl), 150 μL of hydrochloric acid (HCl), 10 mL of urea, 18 mL of calcium chloride dehydrate ($CaCl_2{\cdot}2H_2O$), 1.8 g of BSA, and 30 g of bile. All chemicals and reagents were of ultra-pure quality and obtained from SIGMA, MERCK, and ACS reagents to minimize the contribution of trace heavy metals from these chemicals.