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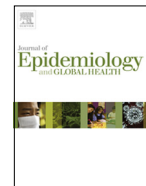
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Drinking water studies: A review on heavy metal, application of biomarker and health risk assessment (a special focus in Malaysia)

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Abstract Malaysia has abundant sources of drinking water from river and groundwater. However, rapid developments have deteriorated quality of drinking water sources in Malaysia. Heavy metal studies in terms of drinking water, applications of health risk assessment and bio-monitoring in Malaysia were reviewed from 2003 to 2013. Studies on heavy metal in drinking water showed the levels are under the permissible limits as suggested by World Health Organization and Malaysian Ministry of Health. Future studies on the applications of health risk assessment are crucial in order to understand the risk of heavy metal exposure through drinking water to Malaysian population. Among the biomarkers that have been reviewed, toenail is the most useful tool to evaluate body burden of heavy metal. Toenails are easy to collect, store, transport and analysed. This review will give a clear guidance for future studies of Malaysian drinking water. In this way, it will help risk managers to minimize the exposure at optimum level as well as the government to formulate policies in safe guarding the population. © 2015 Ministry of Health, Saudi Arabia. Published by Elsevier Ltd. This is an open access article under the CC BY-NC-ND license (<http://creativecommons.org/licenses/by-nc-nd/4.0/>).

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1. Introduction

Malaysia uses 99% water supply for domestic use from surface water, while another 1% of the supply from groundwater [1]. Total internal Malaysia water resources are estimated about 580 km³/year and 30% water withdrawal is for municipal uses [2]. Water supply mainly from surface water and groundwater was treated and distributed to consumers as tap water, bottled drinking water and bottled mineral water which were used as drinking water [3–5]. In Malaysia, main sources of drinking are tap water, bottled drinking water and bottled mineral water [1,6–8]. Water supply from surface water is widely used as drinking water in Malaysia, such as water withdraw from Sungai Langat, Sungai Selangor, Sungai Kinta in West Coast Peninsular Malaysia [3,9]. Water supply from groundwater is also used as drinking water in a few states of Malaysia such as Kelantan, Terengganu, Pahang, Perlis, Kedah, Sabah, and Sarawak [10].

Municipal water consisting of untreated surface water and groundwater needs to be treated, before the water is made potable. A total of 488 water treatment plants (WTP) are operated in Malaysia to treat municipal water before the water is supplied to consumers [11]. Treatment plants in Malaysia have the ability to produce 15,536 Million Litre per Day (MLD) drinking water to consumers [11]. Majority of water treatment plants are using conventional water treatment system, while only a few water treatment plants are using advanced technologies such as Actiflo Clarification System, Ultra Membrane Filtration, Dissolved Air Floatation (DAF) and Ozone [3,12]. Conventional water treatment is divided into three stages namely pre-treatment, pre-chlorination and post-treatment [13]. Pre-treatment stage includes filtration and aeration process to remove particles such as sands, colour, odour and taste [13]. Pre-chlorination phase is functioned to remove smaller particles by pre-chlorination, coagulation (use alum), flocculation (use polymer), sedimentation

and filtration (rapid sand gravity) process [3,13]. Among water treatment plants which are using conventional water treatment systems are at Langat Batu 10 and Cheras Batu 11 WTP (Selangor), Kelar and Kampung Puteh WTP (Kelantan) and Ulu Kinta and Hilir Perak WTP (Perak) [3,9,12]. Usage of advanced technology such as Actiflo Clarification System and Dissolved Air Floatation is to improve clarification process which is similar to coagulation and sedimentation purpose in conventional system [3]. Another advanced technology is Ultra Membrane Filtration which uses transmembrane pressure to remove Cr, Cd, Zn, Cu, Ni and Pb with removal percentage ranging from 92% to 100% [3,14]. Lastly, the post-treatment stage involves disinfection, post lime, fluoride and balancing reservoir to remove bacteria and stabilize water hardness [13]. Ozone technology is used as disinfection to replace chlorination process [15]. List of water treatment plants which are utilized advanced technology are summarized in Table 1. Despite effective heavy metal removal using advanced technologies, small number of this technology used in Malaysia might be due to high technology operation and maintenance with expensive running cost [16].

Treated water from water treatment plant is distributed to consumer as tap water using pipeline system which is provided by water body such as Syarikat Bekalan Air Selangor Sdn. Bhd. (SYABAS) and Syarikat Air Negeri Sembilan Sdn. Bhd. (SAINS) for Selangor and Negeri Sembilan state. Pipeline system used is according to guidelines prepared by National Water Services Commission [13]. There are wide variety of pipes used in Malaysia such as Galvanized Iron (GI), Ductile Iron (DI), Mild Steel (MS), Stainless Steel (SS), asbestos (ABS) and plastics (HDPE and PVC). GI type is the oldest type of pipe that has been used in Malaysia [17]. However, due to low resistant to corrosion, most of states in Malaysia have already replaced this type of pipe with HDPE, SS, DI, and MS pipe [17–19]. In West Coast Peninsular Malaysia and

Table 1 Water treatment plants which are using advanced technology in Malaysia.

State	Reference	Water treatment plant	Water treatment technology
Selangor	[3]	Wangsa Maju Sungai Selangor Phase 2 Sungai Rumpit Kepong	Dissolved Air Floatation (DAF) Actiflo Clarification System Ultra Membrane Filtration
Perak	[9]	Sungai Kinta	Dissolved Air Floatation (DAF)
Kelantan	[12]	Wakaf Bunut Perrala Pintu Geng	Ultra Membrane Filtration Ozone

South Malaysia, all GI pipes have been replaced with HDPE and SS pipe [17,20]. Meanwhile, most of the GI pipes have been replaced by DI and MS pipes in North Malaysia and East Coast Peninsular Malaysia [18,19]. According to WHO, internal corrosion of pipeline can add heavy metals such as Pb, Cu and Fe into drinking water. Furthermore, iron will cause an undesirable taste and colour to the supplied drinking water [21]. The most high resistant material towards corrosion is plastic type due to the polymer material [21]. Plastic pipe such as PVC is widely used as plumbing system at home [17–20]. Another type of pipe that is resistant to corrosion is ABS which is usually used at the coastal area [19,22].

Factors associated with heavy metal contamination in drinking water are the source of drinking water, leaching of the heavy metal from corroded pipeline and unhygienic drinking water practice [23–26]. Most of drinking water sources are from surface water and groundwater which are susceptible to heavy metal pollution due to natural occurrence and anthropogenic activities [27,28]. Heavy metals such as Fe, As, Al, naturally occur in soils that will infiltrate into the water body [28]. Anthropogenic activities such as mining, industrial and agricultural activity also contribute to heavy metal pollution in the water body due to improper wastewater management and run off from fertilizer [26]. However, most of heavy metals from surface water and groundwater are usually removed during water treatment process [24,29]. Furthermore, Malaysian Ministry of Health had listed heavy metal parameters in National Drinking Water Quality Standard to be complied by water body to ensure drinking water supplied is safe for consumers [30]. Nevertheless, heavy metals such as Fe, Pb and Cu could be leached out from the corroded pipeline system [24,31]. Corrosion of pipe occurs resulting from ageing and pitting [24,32]. Pipe lifetime depends on the pipe material and protective lining of inner pipe usually degraded over time which will cause corrosion when water contacts with metal

coating [24]. Pitting is a pin-like formation in the metal surface that is in direct contact with water, will cause the pipe to corrode [32]. Due to this fact, it is a good practice to install private water filters at home to avoid heavy metal contamination [7]. The other factor is unhygienic practices at home such as improper drinking water storage container and unhygienic handlers [25].

Chiron et al. [33] stated that contamination of heavy metal in drinking water is a public health concern due to their absorption and accumulation in humans. Furthermore, heavy metals such as cadmium can displace essential minerals such as vitamin C and E from their metabolically active site and can be toxic to the cell [34,35]. According to Housecroft and Sharpe [36], heavy metals such as Mn, Fe and Ni are needed in enzyme activity. However, heavy metals will be toxic to humans if the metals are exposed or ingested in larger amounts [37–39]. Humans are exposed to heavy metals by ingestion, absorption and inhalation pathways. Ingestion route is indirect exposure by intake of food and drinking water into the gastrointestinal tract [40,41]. Absorption and inhalation routes are direct contact of heavy metal through skin and the respiratory tract in which the aerosol or vapour metal in the air is inhaled into the lung [40,41]. Exposure through the skin is less important than the inhalation and ingestion pathways as discussed by Beckett et al. [40]. Cornelis and Nordberg [41] stated that intake of heavy metal through inhalation pathway is usually small compared to ingestion pathway. The order of ranking for heavy metal exposure routes is absorption, inhalation and lastly ingestion. Thus, ingestion of drinking water is the major source of heavy metal exposure [23,42,43].

2. Objective

This review attempts to guide crucial sources of information on heavy metal concentration status in Malaysian drinking water. Application of HRA in

heavy metal drinking water studies was also assessed in this study. This review also aims to review the most useful tool as a biomarker to be incorporated in heavy metal drinking water studies. An overall brush up of Malaysian drinking water is crucially need to give a clear picture and direction of future innovative and exploration of drinking water studies with potential health risks in Malaysia.

3. Methods

This review included a search of the online electronic databases PUBMED, SCIEDIRECT and Google Scholar. Each database was searched through March to September 2013. Key words used in the search are: drinking water, heavy metal, health risk assessment, biomarker, bio-monitoring, drinking water quality, Malaysia and exposure. Relevant references from the bibliographies of identified papers were searched. Data of heavy metal concentration in drinking water are also extracted from relevant articles and dissertations. These data are standardized in $\mu\text{g/L}$ unit. Importantly, all the methodologies and quality controls for the summarized studies can be viewed from original sources.

4. Results

4.1. Heavy metal in Malaysian drinking water

From a survey conducted by Azlan et al. [8] in Klang Valley showed that 73% respondents consumed drinking water supplied by SYABAS. Studies by Aini et al. [7] and Azlan et al. [8] indicate that drinking water quality is not satisfactory for the consumers. Heavy metal concentrations in drinking water samples are summarized in Table 2. The findings were compared with standards from Malaysian Drinking Water Quality Standard as suggested by Malaysian Ministry of Health (MMOH) [30] and Drinking Water Quality Standard by World Health Organization (WHO) [44]. Most of heavy metal concentration in drinking water were complied with standard limits except for Pb and Al [1,10,45–49]. High concentration of Pb in Bandar Sunway drinking water might be due to source of drinking water which is originated from Selangor River that contain Pb in range of 0.1–50 $\mu\text{g/L}$ [50]. Source of Pb in Selangor River was likely due to anthropogenic sources such as industrial and municipal effluent [50]. Another possible source of Pb in Bandar Sunway drinking water was leaching of the metal from corroded plumbing system [46]. This

can be observed in studies involving comparison between Pb concentrations in the first and fully flushed drinking water which found that Pb in first flush was higher than in the fully flushed tap water [47–49]. The main reason for this difference was corrosion of plumbing system and stagnation of water in the pipes which allow contacts between water with leached Pb [24,47–49,51]. Studies in residential areas in Terengganu, Negeri Sembilan, Perak, Pahang and Johor revealed that Al concentration in drinking water were above the standard limits of MMOH and WHO [52–56]. The findings showed that lack of optimization in coagulation and filtration system of the water treatment are reasons of Al residual in drinking water [29,55,56]. Aluminium sulphate has been used as coagulation and flocculation agent at water treatment plants in the state of Terengganu, Negeri Sembilan, Perak, Pahang and Johor [51–55]. On top of that, the use of excess dose of aluminium sulphate to remove high contaminated organic matter and turbidity in raw water treatment process was also the reason for high Al residual in drinking water [29,51,52,54,56].

According to Jamaludin et al. [60], population in few states such as Kelantan and Sabah who have limited surface water supply are depending on groundwater as the main source for domestic use as well as drinking water. In Kelantan state, 70% of water supply was derived from groundwater [61]. Air Kelantan Sdn. Bhd. (AKSB) has reported that a total of six water treatment plants in Kelantan state are using groundwater sources [62]. Heavy metal levels in groundwater are summarized in Table 3. Most of the studies reported that heavy metal concentration in groundwater was under permissible limit set by Malaysian Drinking Water Quality Standard except for groundwater from Rosob Village, Ampar Tenang and Machang [63–65]. Well water samples from Rosob Village which used as drinking water contain high Mn concentration of 409.5 $\mu\text{g/L}$ compared to 100 $\mu\text{g/L}$ of MMOH permissible limit [64]. According to Kato et al. [64], long term exposure to Mn is associated with neural diseases such as Parkinson disorders. Islami et al. [65] showed that Al concentration in groundwater from Machang violated standard limits with the concentration of 266.2 $\mu\text{g/L}$. High Al concentration in this study site might be due to the agricultural activity as it is located in a palm oil plantation area [65,66]. Rahim et al. [64] reported high concentrations of Fe (1830 $\mu\text{g/L}$), Ni (105.7 $\mu\text{g/L}$), Cd (29.7 $\mu\text{g/L}$) and Pb (505.5 $\mu\text{g/L}$) in the groundwater from Ampar Tenang. The levels exceeded the standard

Table 2 Heavy metal concentration in tap water samples.

Ref.	Mean heavy metal concentration (µg/L)										Instrument
	Range of heavy metals analysed (µg/L)										
	Al	Fe	Cu	Zn	Cr	Mn	Ni	As	Cd	Pb	
[1]	—	59.58 20–330	85.42 10–260	37.41 2–360	0.01 BDL*–0.88	30.9 BDL*–90	0.91 0.26–2.63	0.81 0.16–6.14	0.41 0.36–0.85	0.28 BDL*–3.8	F-AAS* & GF-AAS*
	—	5	1.5	1.5	—	1.5	0.07	—	—	—	*LOD ¹
	—	—	—	—	0.004	—	—	0.05	0.002	0.05	*LOD ²
[10]	—	34.7 11.6–98.7	0.794 0.099–2	130.2 2.2–453	1.24 0.036–3.57	1.43 BDL*–5.33	1.18 BDL*–4.71	—	1.33 0.12–2.56	0.75 0.1–6.03	ICP-MS*
	—	0.0005	0.0002	0.0007	0.0003	0.0001	0.0002	0.0004	0.00007	0.00004	LOD ³
[45]	—	70 23.2–330	8.59 0.9–26.3	34.7 2.1–358.2	—	25.39 BDL*–91	—	—	—	0.32 BDL*–3.8	F-AAS*, GF-AAS* for Pb
	—	5	1.5	1.5	—	1.5	—	—	—	0.05	*LOD ^{1,2}
[46]	—	—	700.0 600–1300	130 30–150	15.0 10–24	—	—	—	0.175 0.1–0.26	32 30–60	*AAS ^x
[52]	206.3 27–611	—	—	—	—	—	—	—	—	—	GF-AAS*
[53]	292.0 53–1030	—	—	—	—	—	—	—	—	—	GF-AAS*
[54]	330.0 10–860	—	—	—	—	—	—	—	—	—	GF-AAS*
[55]	395.5 31–962	—	—	—	—	—	—	—	—	—	GF-AAS*
	0.1	—	—	—	—	—	—	—	—	—	*LOD ²
[56] a*	200.0 130–230	—	—	—	—	—	—	—	—	—	Lambda 25 UV/Vis
[56] b*	220.0 140–360	—	—	—	—	—	—	—	—	—	
[57] c*	110 20–280	—	—	—	—	—	—	—	—	—	Lambda 25 UV/Vis
[57] d*	120 50–260	—	—	—	—	—	—	—	—	—	
	2.0	—	—	—	—	—	—	—	—	—	*LOD ⁴
[58] e*	1.55 0.63–3	—	—	—	—	—	—	—	—	—	GF-AAS*
[58] f*	1.25 0.78–2.09	—	—	—	—	—	—	—	—	—	

(continued on next page)

Table 2 (continued)

Ref.	Mean heavy metal concentration ($\mu\text{g/L}$)										Instrument
	Range of heavy metals analysed ($\mu\text{g/L}$)										
	Al	Fe	Cu	Zn	Cr	Mn	Ni	As	Cd	Pb	
[59] g*	43.59	—	—	—	—	—	—	—	—	—	GF-AAS*
[59] h*	4.97–297	—	—	—	—	—	—	—	—	—	
[47] 1st*	1.02	—	—	—	—	—	—	—	—	—	*LOD ²
[47] 2nd*	0.22–1.83	—	—	—	—	—	—	—	—	—	GF-AAS*
[48] 1st*	0.1	—	—	—	—	—	—	—	—	3.041	
[48] 2nd*	—	—	—	—	—	—	—	—	—	0.09–56.49	
[49] 1st*	—	—	—	—	—	—	—	—	—	1.064	
[49] 2nd*	—	—	—	—	—	—	—	—	—	BDL ¹ –5.22	
[30]	—	—	—	—	—	—	—	—	—	3.22	GF-AAS*
[44]	200	300	1000	3000	50	100	20	10	3.0	0.09–16.6	
[49] 1st*	—	—	—	—	—	—	—	—	—	2.02	
[49] 2nd*	—	—	—	—	—	—	—	—	—	0.05–9.92	
[30]	200	300	1000	3000	50	100	20	10	3.0	0.02–20.52	GF-AAS*
[44]	200	300	1000	3000	50	50	20	10	3.0	0.55	
	—	—	—	—	—	—	—	—	—	0.02–4.62	
	—	—	—	—	—	—	—	—	—	0.05	*LOD ²

* a = Mukim Parit Lubok, Village b = Parit Raja Village, c = Sungai Lembing, d = Bukit Ubi, Village, e = Sungai Michu Village, f = Sungai Buah Village, g = Parit Haji Ibrahim Village, h = Parit Sarang Buaya, 1st = first flush sample and 2nd = sample taken after 2 min flushing [48] and 3 min flushing [49,50], BDL = below detection limit, AASX = Atomic Absorption Spectrometry (type of AAS is unavailable in the literature) F-AAS = Flame Atomic Absorption Spectrometry, ICP-MS = Inductively coupled plasma mass spectrometry, GF-AAS = Graphite Furnace Atomic Absorption Spectrometry, LOD¹ = limit of detection for F-AAS, LOD² = limit of detection for GF-AAS, LOD³ = limit of detection for ICP-MS, LOD⁴ = limit of detection for UV/VIS spectrometry.

Table 3 Heavy metal concentration in groundwater samples.

Ref.	Mean heavy metal concentration (µg/L)										Instrument
	Al	Fe	Cu	Zn	Cr	Mn	Ni	As	Cd	Pb	
[63]	4.0	6.4	1.1	22.4	0.2	409.5	3.0	5.7	BDL*	BDL*	ICP-MS
	0.6–28.1	0.8–47.4	0.2–3.0	1.1–109.7	0.1–1.2	0.2–4000	1.0–7.3	0.2–22.8			
[64]	0.0004	0.0005	0.0002	0.0007	0.0003	0.0001	0.0002	0.0004	0.00007	0.00004	LOD ¹ (µg/L)
	–	1830	550.5	286	19.8	50	105.7	0.35	29.7	505.2	ICP-OES
		950–2212	210–1053	15–820	1–37	40–60	6–328	0.1–2	2–67	138–1107	
[65]	266.2	0.1	0.4	0.2	0.2	0.1	0.5	1	0.1	1	LOD ² (µg/L)
	BDL*	164.7	–	–	–	–	–	–	–	–	ICP ^x
[30]	200	300	1000	3000	50	100	20	10	3.0	10	
[44]	200	300	1000	3000	50	50	20	10	3.0	10	

* BDL = below detection limit, ICP-MS = Inductively coupled plasma mass spectrometry, ICP-OES = Inductively coupled plasma optical emission spectrometry, ICPX = Inductively coupled plasma (type of ICP is not available in the literature), LOD¹ = Limit of detection for ICP-MS, LOD² = Limit of detection for ICP-OES.

limit as the study was conducted near an open dumping area. Thus, the leachate is suspected as the main source of Fe, Ni, Cd and Pb.

Bottled mineral water is referred to as natural mineral water extracted from groundwater, hygienically filtered and bottled [4,5]. Bottled drinking water is water from groundwater, river or drinking water, treated using distillation, reverse osmosis or other suitable techniques and bottled [4,5]. The main difference between bottled mineral water and bottled drinking water is the source of bottled mineral water is groundwater only [4,5]. A bottled water company in Malaysia, Spritzer [67] reported that there is an increased sale of bottled water in 2012 compared to 2011. This trend has shown an increasing demand of bottled water by Malaysian population. The reasons were safety, health, quality and taste [7,8]. Studies on bottled mineral water and bottled drinking water are summarized in Table 4. The studies showed that all the heavy metal concentrations were below the permissible limit of Malaysian Drinking Water Quality Standard which indicated that water is safe to be consumed as drinking water [45,68,69].

4.2. Biomarkers used to monitor human exposure of heavy metal in drinking water

Heavy metal in drinking water studies which have incorporated biomarkers to examine the heavy metal exposure via drinking water were carried out in Chile, Bangladesh, China and the United States [42,70–72]. These biomarkers such as blood, urine, hair and nails [73–82] can reflect body burden of the heavy metal [83,84]. Table 5 shows the biomarkers used to monitor heavy metal in drinking water. However, from the existing literature, there are no studies in Malaysia which have incorporated biomarkers in drinking water studies.

Heavy metals can be determined from the blood components (cells, plasma and serum) and total blood [73,74,85]. Heavy metal in blood reflects 2–3 h exposure, thus making it a good biomarker for recent high dose exposure [74,75]. For example, heavy metal in blood would be suitable to study exposure of high heavy metal concentration in drinking water in countries such as Bangladesh [75]. However, Marchiset-Ferlay et al. [74] concluded that blood is not an ideal biomarker for heavy metal exposure in drinking water, however, can be used as a comparison with other biomarkers. A few studies have observed correlations between heavy metal concentration in urine, blood and water [70,76]. The main limitation of this biomarker is the invasive collection method which may reduce participation rate from the population.

Table 4 Heavy metal concentration in bottled mineral water (MW) and bottled drinking water (BW) sample.

Ref.	Bottled water type	Mean heavy metal concentration ($\mu\text{g/L}$)									Instrument
		Range of heavy metals analysed ($\mu\text{g/L}$)									
		Fe	Cu	Zn	Cr	Mn	Ni	As	Cd	Pb	
[45]	MW	11.62	12.77	4.79	—	31.54	—	—	—	0.26	F-AAS [*] for Fe, Cu, Zn & Mn,
	BW	BDL [*] –60.5	1.5–16.9	0.4–24.3	—	BDL [*] –68	—	—	—	BDL [*] –1.25	
		34.51	2.99	1.31	—	5.14	—	—	—	1.28	GF-AAS [*] for Pb
		17.2–43.3	0.2–7.3	BDL [*] –2.7	—	BDL–31	—	—	—	BDL [*] –3.92	
[68]	MW	11.55	12.77	4.79	BDL [*]	31.53	1.5	3.2	0.36	0.26	F-AAS [*] for Fe, Cu, Zn, Cr & Mn,
	BW	BDL [*] –60.5	1.5–16.9	0.4–24.3	—	BDL [*] –68	0.28–6.88	BDL [*] –7.7	0.3–0.45	BDL [*] –1.25	
		34.51	2.99	1.19	0.4	5.14	0.55	0.38	0.49	1.28	GF-AAS [*] for Ni, As,
		17.2–43.3	0.2–7.3	BDL [*] –2.7	BDL [*] –2.56	BDL–31	0.13–2.09	BDL [*] –1.68	0.38–0.85	BDL [*] –3.92	Cd & Pb
		5	1.5	1.5	3	1.5	—	—	—	—	*LOD ¹ ($\mu\text{g/L}$)
							0.07	0.05	0.002	0.05	*LOD ² ($\mu\text{g/L}$)
[69]	MW	—	0.64	11.5	—	—	—	—	—	—	ICP-MS [*]
	BW	—	0.11–1.71	0.96–41.3	—	—	—	—	—	—	
		—	0.24	6.20	—	—	—	—	—	—	
		—	0.03–0.96	0.19–40.5	—	—	—	—	—	—	
		—	0.0002	0.0007	—	—	—	—	—	—	*LOD ³ ($\mu\text{g/L}$)
[30]		300	1000	3000	50	100	20	10	3.0	10	
[44]		300	1000	3000	50	50	20	10	3.0	10	

* BDL = below detection limit, F-AAS = Flame Atomic Absorption Spectrometry, GF-AAS = Graphite Furnace Atomic Absorption Spectrometry, ICP-MS = Inductively coupled plasma mass spectrometry, LOD¹ = limit of detection for F-AAS, LOD² = limit of detection for GF-AAS, LOD³ = limit of detection for ICP-MS.

Table 5 Summarization of biomarkers used in drinking water studies and their correlation with heavy metals in drinking water.

References	Samples	Main result
[42]	Urine	As water intake is the main source of concentration of As in urine
[70]	Water	
	Blood	(1) Positive association between As in urine and plasma folates ($r = 0.14$)
	Urine	(2) Negative association between As in urine and total homocysteine ($r = -0.14$)
	Water	
[71]	Urine	Increasing of As in urine is correlated with increasing As concentration in water
[72]	Water	
	Urine	Correlation between As in:
	Toenail	(1) Tap water and urine ($r = 0.35$)
	Water	(2) Tap water and toenail ($r = 0.33$)
		(3) Urine and toenail ($r = 0.36$)
[75]	Blood	As in water is more correlated to As in toenail ($r = 0.36$) than urine ($r = 0.017$)
	Urine	
	Toenail	
	Water	
[76]	Blood	Correlation between:
	Urine	(1) Blood As and water ($r = 0.76$)
	Water	(2) Urinary As and water ($r = 0.76$)
		(3) Blood As and urinary As ($r = 0.85$)
[77]	Urine	Correlation between As in:
	Hair	(1) Fingernail and water ($r = 0.48$)
	Fingernail	(2) Hair and water ($r = 0.48$)
	Blood	(3) Urine and water ($r = 0.75$)
	Water	
[86]	Urine	As concentration in urine from exposed group is higher than control group
	Water	
[88]	Toenail	Significant correlation between As in toenail and groundwater ($r = 0.84$)
	Water	

According to Beckett et al. [40], urine was the main route of elimination for many heavy metal such as As, Cd, Cr, Cu, Ni, and Zn. Heavy metal in urine also has short residence time of 3–4 days which shows current exposure [75,77,85]. Studies conducted in Chile, China and USA indicated correlations between heavy metal in urine and drinking water [42,71,72,86]. There are two types of urine sampling methods which are spot sample and timed urine sample [73]. Aitio et al. [73] stated that spot samples are usually taken because it is simple compared to timed urine sample which requires detailed instruction, training and supervision. Interpretation of heavy metal in spot sample needs urine creatinine adjustment which depends on demographic factor rather than analytical technique [87]. Urine samples must be analysed as soon as possible after sampling because storage may alter or reduce the concentrations of the heavy metal species [74,86]. On top of that, Nermell et al. [87] concluded that heavy metal in urine reflects the excretion despite actual body burden.

Another non-invasive biomarker is hair. Disulphide bonds in hair keratin allow heavy metal

retention [89,90]. Hairs also accumulate a wide range of heavy and trace metals such as Cd, Pb, Cr, Cu, Al, Fe and Ni [83,91]. Elements in hair are detached from metabolic processes after the formation and make it a stable marker for long term exposure [90,92]. The main limitation of this biomarker is the exposure to external contaminants such as shampoo, dust and cosmetic procedures (bleaching, dyeing and permanent waving) [80,93]. Mandal et al. [77] have conducted an experiment to understand adsorbing characteristic of hair and fingernail. The study found that adsorption of heavy metals on hair surface is a critical problem in epidemiology studies while adsorption of heavy metals on fingernail is negligible [77]. Furthermore, a study by Harkins and Susten [93] showed that hair also have high variable in inter-individual growth rates.

Thus, incorporation of biomarkers in Malaysian drinking water studies should consider toenail as a useful tool. This is because toenail is a useful tool in bio-monitoring compared to blood, urine and hair due to their rapid growth, less external contamination, adequate sample availability and

incorporation of elements in the tissue [34,94]. Nails are metabolic end product of skin that reflects the element composition of cells [34]. According to Adair et al. [75], toenail was a useful tool in exposure assessment study because they were less exposed to water than fingernail, skin and hair. Toenails provide longer time for heavy metal accumulation due to slower growth rate compared to fingernail [78]. Toenails are easier to collect the sample, store, transport and prepare for analysis [78,80]. He et al. [28] stated that ultrasonic cleaning using polar and non-polar solvent is efficient to remove external heavy metal contamination of toenail including nail polish without altering heavy metal content in the toenails. Furthermore, adsorption of external heavy metal on nail surface is negligible compared to hair [77]. On the other hand, Hinwood et al. [95] showed that inter-individual variability of toenails was lower than hair. According to Slotnick and Nriagu [90], inter-individual variability of toenail growth was associated with climate. Toenail growth among people is different during warm and cold climate [90]. Toenail growth is faster in summer compared to winter [90]. Due to faster growth rate during summer, heavy metal concentration in toenail should be lower in summer compared to winter [78,90]. However, Karagas et al. [72] has found that heavy metals in toenail were slightly higher during summer. The finding shows that difference in growth rate does not affect heavy metal concentration in toenail [90,72]. The heavy metal concentration difference in toenail is due to changes in drinking water consumption which is higher during summer than winter [72]. Analysis of blood and urine reflects current exposure of 2–3 h and 3–4 days respectively while toenail indicates exposure of 2–12 months before sample collection [75,79,80,90]. Due to this fact, assessment of heavy metals in drinking water used by target population must be done at least 2 months before toenail collection to ensure that heavy metals analysed in the toenail were represented by the drinking water exposure. Heavy metals accumulated in the toenail are considered as stable over time since nails that detached from the skin were isolated from other metabolic activities in the body [95]. Few studies have reported a positive correlation between concentrations of heavy metal in drinking water and toenail [72,75,91].

There were a few confounders which contribute to heavy metal in these biomarkers. The main confounder is dietary intake of seafood [74,96–98]. Other important confounders were dietary habit,

body size, age, sex, smoking habit, health conditions, medicine and supplement intake [88,95,98,99]. Nevertheless, exposure of heavy metal is more significant through ingestion routes while dermal exposure was negligible for relatively low heavy metal water such as in Malaysia [40]. Razak et al. have concluded that confounder information can be collected using questionnaires in drinking water study to understand the relationship of ingestion exposure and heavy metal reflected by biomarker [100]. Studies of drinking water exposure incorporating biomarkers need to be designed carefully at early stage or later using robust statistical methodology to ensure that heavy metal in the biomarker was reflected by drinking water exposure and does not affected by confounders and other routes of exposure [90,94,100]. Any bio-monitoring programme or research to ascertain body burden of heavy metal through drinking water must consider all these confounders to avoid false interpretation of heavy metal source.

4.3. Health risk assessment (HRA) application in drinking water studies in Malaysia

HRA is crucial to understand the potential health risk from the heavy metal exposure to humans [55,101,102]. This information is very important for decision makers to set up policies or regulations to protect population's health [103,104]. Only few drinking water studies in Malaysia have applied HRA in their studies [51,52,55–57]. HRA of heavy metal ingestion pathway through drinking water may not receive attention in Malaysia due to the low levels of heavy metal in drinking water [10,45,55,56,67]. However, high rate of drinking water intake by population could increase the risk of exposure since intake rate is one of the variables in the HRA calculation [102].

HRA of Pb exposure was carried out in particular housing areas of Selangor, Terengganu and Negeri Sembilan [47–49]. These studies showed that there were no potential adverse effects from Pb intakes via drinking water. Studies done in Terengganu, Negeri Sembilan, Pahang, Selangor and Johor found that there were no potential adverse effects from Al intake in drinking water [52–58]. However, Nora [59] has found that only 6.7% of the population from Parit Haji Ibrahim Village, Johor has hazard index of more than 1 which indicated that the population was at risk of getting Alzheimer and Parkinson diseases by consuming high Al concentrations in drinking water.

Currently, there is a rising interest of biomarker application in HRA studies [104,105]. In addition, Sobus et al. [104] stated that biomarker is the most appropriate tool to represent effect of exposure from the source to health outcome. Toxicity data from high-dose animal toxicity test and screening of exposure levels resulted in high uncertainties of association between exposure and health outcome [104]. Thus, application of biomarker in HRA can provide a better assessment since the tool is more specific and sensitive than most of clinical tests [104,105]. Studies by Egeghy et al. [106] and Thomas et al. [107] showed that estimated exposure of pollutants can be supported by measurement of the pollutants in biomarkers.

5. Conclusion

The review has highlighted findings of heavy metal in drinking water from studies carried out in Malaysia. The summary of these studies concluded that heavy metal in Malaysian drinking water are still under permissible limits. Biomarkers of heavy metal exposure in drinking water such as blood, urine, hair and toenail have been used in drinking water studies elsewhere. Among all the biomarkers, toenail appears as the most useful tool to monitor accumulation of heavy metals in the human body. This is due to the fact that toenails are less contaminated, their rapid growth can provide adequate sample and binding of heavy metal in the tissue. The HRA application in heavy metal ingestion study in Malaysia is also still limited. HRA is important to be included in drinking water studies because it can be used to estimate the potential of adverse health effects in humans. Incorporation of biomarker in future drinking water studies is crucial to fill up the knowledge gap of heavy metal accumulation in Malaysian population. Future studies including HRA are vital in understanding the risk of heavy metal exposure in drinking water and help government as well as risk managers to find ways to minimize the exposure.

Conflict of interests

The authors declare that there are no conflicts of interest.

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