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Evaluation of Industrial Workplace Exposure to Metal Fumes using Toenail as Bio-Indicator

Siti Farhana Zainal Bakri[1,2*], Azian Hariri[2], Marzuki Ismail[3], Amir Abdullah Muhamad Damanhuri[4]

[1]DRB-HICOM University of Automotive Malaysia,
Faculty of Engineering and Technology, Pekan, 26607, MALAYSIA

[2]Universiti Tun Hussein Onn Malaysia,
Faculty of Mechanical and Manufacturing Engineering, Batu Pahat, 86400, MALAYSIA

[3]Universiti Malaysia Terengganu,
School of Ocean Engineering, Kuala Terengganu, 21300, MALAYSIA

[4]Universiti Teknikal Malaysia Melaka,
Faculty of Engineering Technology Mechanical and Manufacturing, Durian Tunggal, 76100, MALAYSIA

*Corresponding Author

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Abstract: The toxic gaseous generated from welding process contained heavy metals that induce various cancers and diseases. Exposure to this toxic fume is associated with the occurrence of lung cancer and worker that exposed to this situation is considered as a high-risk group. This study aims to investigate the amounts of heavy metals concentration in the breathing zone and toenail samples of 36 individuals from automotive related industries in Malaysia that were classified as an exposed group (welders) and non-exposed group (office workers) by cross-sectional research design. Personal air sample and mixed cellulose ester (MCE) filters were used to collect the welding fumes while toenail samples from all 10 toes of each subject for biomonitoring. Collected samples were digested, and analyzed to evaluate the levels of heavy metals by inductively coupled plasma mass spectrometry (ICP-MS). Eleven heavy metals (Al, Cr, Mn, Fe, Co, Ni, Zn, Cu, As, Ag, and Pb) were measured in all samples; and results showed that the concentrations of Cr, Zn, As, Ag and Pb were significantly difference ($P \leq 0.05$) for exposed and non-exposed groups. Furthermore, the metal fume exposure generated from different welding types in these three plant shows a similar exposure to the welders. Thus, more attention need to be considered to the working conditions of the exposed group, specifically on providing proper ventilation at the workplace as well as to provide a suitable protective personal devices.

Keywords: Carcinogenic metal, toenail biomarker, welding fumes, Malaysia.

1. Introduction

Heavy metals are a group of metals and metalloids that normally present naturally, released into the environment by both natural and anthropogenic sources. These are toxic in any state for humans and animals even at low concentration [1]. Most of the non-biodegradable elements are classified as human carcinogenic (known or probable) according to the International Agency for Research on Cancer (IARC) who have a tendency to accumulate in a living organism and may cause adverse health effects either in the long term or continuous exposure [2]. In recent years,

*Corresponding author: farhana@dhu.edu.my

investigating heavy metal is a continuing concern within global public health. The human exposure has increased dramatically because of a few factors as a result of the heavy metals application in industrial process and product. The metal-based operation in industries such as welding is a point source area that makes the workplace environmental pollution became very prominent. Excessive exposure to heavy metals can lead to a variety of disease and adverse effects due to cellular and tissue damage [3]–[5]. The toxicity of heavy metal can damage the function of body systems and important organ such as lungs, brain, liver, kidney and also can cause neurological degenerative processes [6]. This element able to disturb the important biochemical processes that lead to oxidative damage and has the potential to cause chronic inflammatory disease and cancer [7]. Generally, the heavy metal may enter the human body through inhalation of dust, direct ingestion of soil, water or consumption of vegetable from contaminated fields as well as dermal absorption [8]. However, in an occupational setting, inhalation is the common route of exposure to human. To date, little studies have examined the association between heavy metals concentration in the workplace and human body-using toenail as a biomarker. Thus, the aim of this paper is to explore the relationship between the existences of heavy metal concentrations in the workplace environment with heavy metal found in the worker’s body.

2. Methodology

2.1 Study Population

The target population of this study is among Malaysian male welders with at least one year of experience as a welder (exposed group) and administrative worker who worked in the same industries and did not expose to any welding fumes (non-exposed group) in Peninsular Malaysia. The entire involved participant comes from three different automotive-related plants located in both Malacca and Pahang areas. These industries have the same business that related to the manufacture and assembly of the automotive-related component. Participants were recruited between October 2016 and April 2018 with a ration of the non-exposed group and exposed group were approximately 1:3. A total of 36 participants have provided biological samples: 27 participants in the exposed group and nine participants (eight males and one female) in the non-exposed group. All study subjects provided toenail samples to assess the body concentrations of heavy metals. Overall, these workers had normal working hours, with the average exposure among welders is 8hr/day or 40hr/week.

2.2 Sample Collection and Analysis

Samples are collected from each factory to determine a welding fume concentrations in the welding areas by using a personal air pump Gillian 5000 (Sensidyne, USA) with three-piece sampling cassette (SKC Ltd, UK) that equipped with 37 mm 0.8 µm mixed cellulose ester (MCE) filter and supporting media with 1.7 L/min flow rate. The air is collected in the breathing zone in front of the face within 0.2 to 0.3 m diameters from the nostrils of the welder. For biomonitoring, toenail samples were collected from all 10 toes of each subject by using stainless-steel nail clippers and stored in a labelled envelope. Toenail clippings were analyzed by following a modified version of Method 3050B, USEPA as described in previous research [9], [10]. All filter paper and toenail samples were transferred to the accredited laboratory for analysis by using inductively coupled plasma mass spectrometry (ICP-MS) with microwave digestion method according to ASTM D7439-08 at the Central Laboratory, Universiti Malaysia Pahang. The assessed elements in this research are Al, Cr, Mn, Fe, Co, Ni, Zn, Cu, As, Ag, Cd, and Pb.

2.3 Statistical Analysis

The distribution data had positive skewness and are analyzed using IBM SPSS Statistics for Window version 23 (IBM SPSS Inc., Chicago, IL, USA) and Microsoft Office Excels 2016. The descriptive statistics of age, height, weight, and smoking status are calculated. Mann-Whitney test is used to analyze the significance of the differences between the means concentration of heavy metal in toenail of controls and exposed groups. The Kruskal-Wallis test uses to compare the averages of three different welding type as the independent groups (MIG, spot weld and robotic weld) in the plant. The significance of correlations between parameters (toenail metals levels and airborne metals concentrations) is determined by Spearman’s rank correlation coefficient. The interpretation of Spearman’s rho is adapted from Dancey and Reidy (2004) [11]. Statistical significance was defined as a p-value of <0.05.

2.4 Ethical Consideration

The present study approval was taken from a research ethics review committee of International Islamic University Malaysia, Pahang, Malaysia (Ref. ID: IREC 850). Verbal and written informed consent was taken from the participants.

3. Results and Discussion

Globally, researchers have reported the adverse health effects from occupational exposure to welding fumes [12]–[15]. Welders are commonly exposed to carcinogenic metals such as manganese, lead, iron, cadmium, nickel and such that generated by MIG, spot weld and robotic welding [16]–[19]. Frequencies of age, height, weight, and smoking habit of the participant are shown in Table 1. The involved participants are 97% Malay and 3% Indian with the majority is classified as ever-smokers (current and ex-smokers). A former smoker is those who had smoked for at least one year but not during the last 12 months [20].

Table 1: Demographic data of participated welders and control in biomonitoring.

Group	Variable (mean ± SD)			
	Age (yr)	**Height (cm)**	**Weight (kg)**	**Ever-Smokers % (n)**
Exposed (N = 27)	27.44±8.09	1.68±0.08	68.54±21.99	85% (23)
Non-exposed (N = 9)	35.33±11.26	1.66±0.06	64.09±9.79	67% (6)

In overall, the result of heavy metal concentrations in the welding-working environment and the toenail samples are summarized in Table 2. Out of 14-targeted heavy metals, 11 heavy metals were detected by ICP-MS at the welding area samples, while 12 toxic metals are traced in toenail specimens. In the analysed samples, there is no beryllium (Be) and molybdenum (Mo) were detected in both personal sampling and biomarker while none of the cadmium (Cd) elements is traced in three different plants.

Table 2: Summary statistics of heavy metal concentration in the workplace environment (mg/m^3) emitted from different welding type and toenail specimens (ug/g) of exposed and non-exposed worker.

Metal	Toxic Metal Concentration in workplace environment (mg/m^3)									PEL USECHH (mg/m^3)
	MIG			**Spot Weld**			**Robotic Weld**			
	AM	**GM**	**SD**	**AM**	**GM**	**SD**	**AM**	**GM**	**SD**	
Al	44.35	23.81	29.37	54.54	53.42	15.54	48.38	44.92	20.88	5.00
Cr	237.78	188.86	97.38	204.69	196.16	82.70	232.30	223.96	80.27	0.50
Mn	604.25	61.09	1152.74	6.45	6.37	1.40	9.19	9.17	0.88	0.20
Fe	590.69	169.33	780.05	228.86	191.53	177.17	165.08	24.30	184.79	5.00
Co	0.38	0.17	0.26	1.03	0.89	0.72	0.54	0.51	0.22	0.10
Ni	13.46	2.56	26.81	3.29	2.97	2.00	0.89	0.83	0.35	1.50
Cu	8.64	7.42	6.39	5.72	4.86	4.26	4.58	4.29	1.99	0.20
Zn	11.99	6.03	11.60	8.60	5.86	8.90	8.96	7.32	5.62	5.00
As	68.94	38.74	40.01	51.58	50.66	13.68	58.58	58.20	8.38	0.01
Ag	4.73	1.65	6.78	1.62	1.56	0.62	0.43	0.41	0.15	0.10
Pb	77.21	70.46	35.48	63.09	63.09	0.42	61.38	57.21	25.50	0.05

Metal	Toxic Metal Concentration in Toenail (ug/g)					
	Exposed Worker			**Non-exposed**		
	AM	**GM**	**SD**	**AM**	**GM**	**SD**
Al	53.11	13.87	62.24	1025.11	32.78	2338.02
Cr	118.66	52.12	217.60	1257.63	399.02	2100.63
Mn	4.08	1.31	12.39	5.54	1.75	7.64
Fe	115.73	0.61	403.63	40.68	0.22	117.12
Co	1.77	0.01	8.98	0.16	0.05	0.28
Ni	18.66	5.66	21.17	38.78	12.96	45.30
Cu	1.39	0.27	4.95	2228.44	2.06	6681.37
Zn	55.83	47.35	22.23	218.86	133.36	278.04
As	15.32	2.68	27.43	167.23	54.23	281.54
Ag	0.10	0.01	0.39	1.64	0.16	4.24
Cd	0.27	0.02	0.45	4.44	0.36	6.39
Pb	24.67	4.70	65.70	211.47	53.88	257.87

Notes: Abbreviation: PEL: Permissible exposure limit; USECHH 2000: Use and Standards of Exposure of Chemicals Hazardous to Health (USECHH) Regulations 2000; AM: arithmetic mean; GM: geometric mean; SD: standard deviation.

The mean concentration of the heavy metal found in the toenail are listed in the following order: Cr > Fe > Zn > Al > Pb > Ni > As > Mn > Co > Cu > Cd > Ag (exposed group) and Cu > Cr > Al > Zn > Pb > As > Fe > Ni > Mn > Cd > Ag > Co (non-exposed group) respectively. While the mean concentration of heavy metal found in breathing zone is allocated by the following sequences: Mn > Fe > Cr > Pb > As > Al > Ni > Zn > Cu > Ag > Co (MIG); Fe > Cr > Pb > Al > As > Zn > Mn > Cu > Ni > Ag > Co (spot welding); and Cr > Fe > Pb > As > Al > Mn > Zn > Cu > Ni > Co > Ag (robotic welding). The heavy metals in both toenail and cassette samples show the existence of broad range metal concentrations and this were interpreted by large standard deviations in the data samples. Another important finding revealed that all of the average metals concentration generated from the welding activities that related with MIG, spot weld and robotic weld in three plants have exceeded the safe level allowed by USECHH Regulations 2000 except for nickel from robotic weld.

There was a high prevalence of pulmonary diseases in welder's exposure especially to chromium, beryllium, iron, nickel, and aluminium [21]–[24]. In this study, the highest concentration of metal fume is found higher in welder's breathing zone compared to the findings of the other similar study [15], [25]–[27]. Therefore, there is a potential possibility of the welder to experience pulmonary effects due to metal fume exposure in these plants. This exposure may occur due to poor ventilation in welding areas as well as lack of and/or inappropriate wearing of personal protective devices. This study was supported by the previous study where the exposed workers who work near to the point source pose a high risk of heavy metals exposure and have significant heavy metal intoxication even in a low concentration [28].

Toenail specimen, as a recommended biological indicator of heavy metal, were selected to detect the subjects' exposure to 12 types of heavy metals [29]–[31]. The comparison of heavy metal concentrations in toenail from the exposed and non-exposed group is shown in Table 3. Two-independent sample of Mann-Whitney test revealed that there were statistically significant differences ($p < 0.05$) among exposed and non-exposed groups in a trace element of Cr, Zn, As, Ag, Cd, and Pb, except for Al, Mn, Fe, Co, Ni and Cu. The element concentration of aluminium, manganese, iron, cobalt, nickel, and copper that were found in the non-exposed group is not a surprise since 67% of the non-exposed group participants is categorised as ever-smoker.

Table 3: Mann Whitney test of toxic metal concentration (ug/g) in toenail of exposed and non-exposed worker.

Metal	Toenail (ug/g)		
	Mann-Whitney Test	**Z**	***p*-value**
Al	97.0000	-0.895	0.371
Cr	25.000	-3.525	0.001*
Mn	95.500	-0.950	0.342
Fe	113.000	-0.311	0.756
Co	81.000	-1.480	0.139
Ni	98.000	-0.859	0.391
Cu	81.000	-1.480	0.139
Zn	21.000	-3.458	0.001*
As	25.000	-3.526	0.001*
Ag	33.000	-3.234	0.001*
Cd	68.000	-1.958	0.050*
Pb	59.000	-2.283	0.022*

Notes: *Statistically significant difference with $p<0.05$ level.

This habit may influence the number of metals found in the control's body. Despite, it has been suggested that the accumulation of heavy metal in the human body also depends on seafood intake especially shellfish, marine fish and smoking habit [32], [33].

Table 4: Kruskal-Wallis test of heavy metal concentration found in the breathing zone emitted from three different types of welding activities.

Metal	MIG (n)	Spot weld (n)	Robotic weld (n)	Chi-square	df	p-value
Al	9	2	3	0.13	2	0.94
Cr	9	2	3	0.43	2	0.81
Mn	9	2	3	4.96	2	0.08
Fe	9	2	3	0.87	2	0.65
Co	9	2	3	2.70	2	0.23
Ni	9	2	3	3.16	2	0.21
Cu	9	2	3	2.19	2	0.33
Zn	9	2	3	0.04	2	0.98
As	9	2	3	2.97	2	0.23
Ag	7	2	3	2.69	2	0.26
Pb	7	2	3	0.54	2	0.77

Note: Abbreviation: df: the degree of freedom.

This is proven by even though there is no cadmium concentration was traced in any welding activities area, the existence of cadmium in the toenail of both groups may arise due to the smoking habit as well as food intake [34], [35]. The local study has shown that there is the bioaccumulation of toxic metals of Cd, Cu, Fe, Mn, Pb and Zn in fish samples with the highest average concentration found is Fe [36]. The comparison with the previous study has shown that the toenail metal concentration found in welders is lower than these research findings [17], [37]. There are several possible explanations for this result. The discrepancy could be attributed to the welding-type, type of electrode, filler material, base metal and etc as this process has evolved with a wide set of ranging application from deep-sea explorations to the outer space navigations [38], [39]. The data of heavy metal concentration that are emitted from three different welding type are tabulated in Table 4. The current study found that there is no statistically significant difference between the heavy metal discharged from different welding type in three industrial areas. The distribution of metal concentration for three different types of welding has a similar concentration pattern. Thus, the exposure to welding fume among welders in three different plants is assumed to have similar exposure.

The association degree and monotonic relationship of a metal traced element in welder's toenail specimen was measured using Spearman's correlation coefficient analysis and the results are presented in Table 5. This statistical evidence had suggested that the existence of metals in the welder's toenail had a similar anthropogenic source of activity and there are strong significant correlations of toenail metal concentration with one another, with the exception of iron, cobalt, and nickel.

Table 5: Spearman's correlation coefficient (r_s) matrix for pairs of heavy metals found in welder's toenail samples (ug/g).

	Al	**Cr**	**Mn**	**Fe**	**Co**	**Ni**	**Cu**	**Zn**	**As**	**Ag**	**Cd**	**Pb**
Al	**1**											
Cr	-0.48	**1**										
Mn	-0.21	-0.31	**1**									
Fe	0.31	-0.23	-0.02	**1**								
Co	0.19	0.09	-0.48*	0.15	**1**							
Ni	0.38	-0.23	-0.05	0.21	0.13	**1**						
Cu	0.13	0.15	0.56*	-0.04	-0.31	0.07	**1**					
Zn	0.08	0.10	0.39	-0.02	-0.13	-0.00	0.77**	**1**				
As	-0.35	0.82**	-0.33	-0.29	0.32	-0.07	0.16	0.04	**1**			
Ag	0.39*	0.11	-0.13	0.09	0.17	0.32	0.12	0.07	0.28	**1**		
Cd	0.10	-0.11	0.38*	-0.24	-0.28	0.07	0.23	0.03	0.01	0.36	**1**	
Pb	0.12	0.04	0.27	-0.24	-0.16	0.02	0.47*	0.25	0.19	0.44*	0.85**	**1**

Notes:* Correlation is significant at the 0.05 level (2-tailed), and **: Correlation is significant at the 0.01 level (2-tailed).

Correlations between airborne and toenail concentrations for eleven metals were investigated using Spearman's correlation coefficient analysis and the result was bold and expressed in Table 6. Results showed that the correlation coefficients range from 0.11 to 0.59. There was a moderate relationship of silver in toenail and workplace environment, while, a strong relationship is found in manganese, iron, cobalt, and lead between a worker who exposed to carcinogenic metals in automotive-related component industries and their toenail concentrations. The strong negative correlation indicates that the anthropogenic origin source of metal in toenail does correlate with the workplace welding fumes emission.

Table 6: Spearman's correlation coefficient (r_s) in measuring heavy metal found in welder's toenail (ug/g) and heavy metals captured in the workplace environment (mg/m^3).

Metals		**r_s Toenail Specimen (ug/g)**										
		Al	**Cr**	**Mn**	**Fe**	**Co**	**Ni**	**Cu**	**Zn**	**As**	**Ag**	**Pb**
r_s Workplace environment (mg/m^3)	**Al**	**-0.15**	-0.12	0.23	0.14	-0.16	0.10	0.37	0.21	-0.13	0.12	0.41
	Cr	0.04	**-0.11**	-0.21	-0.32	0.30	0.48	-0.03	-0.22	-0.00	0.08	0.24
	Mn	0.31	0.06	**-0.51**	0.02	0.53	0.46	-0.64*	-0.49	0.08	0.11	-0.61*
	Fe	0.06	-0.00	-0.27	**0.49**	0.17	0.02	-0.45	-0.60*	0.02	0.17	-0.24
	Co	-0.27	-0.05	0.62*	0.09	**-0.53***	-0.17	0.59*	0.59*	-0.08	-0.01	0.44
	Ni	0.25	0.10	0.30	0.28	-0.04	**-0.29**	0.17	0.23	0.06	-0.52	-0.20
	Cu	0.20	-0.10	0.10	0.21	0.04	-0.11	**-0.18**	-0.13	-0.06	0.16	-0.27
	Zn	-0.06	-0.18	0.32	0.06	-0.24	0.08	0.31	**0.25**	-0.19	0.19	0.40
	As	0.35	-0.29	-0.17	-0.05	0.30	0.39	-0.28	-0.40	**-0.19**	0.09	-0.06
	Ag	0.29	-0.08	0.27	0.34	0.07	-0.18	0.40	0.30	-0.09	**-0.34**	-0.15
	Pb	0.04	-0.06	-0.21	0.41	0.09	-0.08	-0.27	-0.11	-0.05	0.21	**-0.59***

Notes:* Correlation is significant at the 0.05 level (2-tailed), and **: Correlation is significant at the 0.01 level (2-tailed).

4. Conclusion

In summary, the results indicate that the heavy metals concentration found in the toenail of the exposed and non-exposed group is statistically difference except for Al, Mn, Fe, Co, Ni and Cu. The existence of these constituents in the toenail may portray from the smoking habit as well as possible due to seafood intake. One of the more significant findings from this study is that the metal fume exposure generated from different welding types in these three plant has shown a similar exposure to the welders. Furthermore, a medium to a very strong relationship of chemical concentration was found in both work environment and toenail of heavy metals except for three metals (Fe, Co and Ni). Thus, for any kind of welding type, more attention needs to be considered for conducive working conditions, specifically on having proper ventilation at the workplace as well as providing suitable protective personal devices to the exposed group.

Conflict of Interest

The authors firmly declare that there are no conflicts of interest related to this work.

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