# Heavy Metals (Pb and Cu) Assessments in Hair Samples of Goldsmiths in Kelantan, Malaysia

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### ABSTRACT

**Objective:** This study analyzed the occupational related exposure to lead (Pb) and copper (Cu) in hair samples among goldsmiths in Malaysia.

Method: This cross-sectional study was done among goldsmiths in Kelantan. Background information was obtained and hair samples were collected. Pb and Cu in hair were analyzed via Inductively Coupled Plasma -Optical Emission Spectrometry method.

**Result:** A total of 40 workers from 10 gold workshops were recruited for this study. Most of the respondents were male and Malays aged between 30-39 years old. Two-third of the respondents had diploma level of education with income between RM1000-RM2999. The mean (standard deviation) and geometric mean for Pb was 72.0 (107.3) µg/g and 31.9 µg/g, respectively; whereas for Cu was 35.4 (22.9) µg/g and 28.9 µg/g, respectively. Heavy metal levels were directly associated with overtime work whereas safety and health practice was not linked with protective effect from exposure. Pb and Cu levels were significantly correlated (r=0.36, p=0.023).

Conclusion: Higher hair heavy metal levels were detected among goldsmiths in this study compared to other studies. Longer duration of overtime work was associated with higher heavy metal exposure. Significant positive correlation between Pb and Cu suggest that workers may likely be exposed to metal fumes via occupational sources. Engineering control solutions such as the use of local exhaust ventilation should be placed at work sections to reduce toxic metal exposures and more efforts needed to improve the safety and health practice at gold workshops for a better protection to all workers from such exposures.

Keywords: occupational, biological monitoring, lead, copper, jeweler, metal fumes

# 1. Introduction

containing inorganic chemicals that may be

carcinogenic which occur during the production of gold jewelry. These inorganic chemical exposures may occur during the steps of molding, casting, filling, polishing Goldsmiths are exposed to dusts, fumes and vapors and plating gold jewelries (Choudhari et al, 2013). Inorganic chemicals have been shown to consist of heavy metals and trace elements such as lead (Pb), copper (Cu), cadmium (Cd) and mercury (Hg). The route of exposures to metal fumes, vapors and dusts is largely via inhalation. Most of the exposures tends to accumulate chronically as the goldsmiths are exposed daily at work. Furthermore, under conditions of continued exposure, not all of the metals that enter the body will be eliminated. Instead the exposure will accumulate in body tissues like hair and bone (ATSDR, 2007) and may cause heavy metal toxicological effects and diseases.

Gold-related businesses are common in Malaysia and there are a number of large jewelry maker and retailers at the market presently. In addition to large jewelry retailers, small to medium-sized gold workshops which are usually self-employed and independent and are run by family members or small number of workers has managed to sustain its businesses and thrive in many parts of the smaller town areas in peninsular Malaysia. These small gold workshops offer services to produce and design jewelries according to the demand of their customers.

Unlike established manufacturers of gold jewelry, small gold workshops may neither have the expertise in safety and health nor have adequate resources to provide effective engineering control methods to reduce workers exposures to metal fumes and dusts. These metal fumes and dusts can be originated from the jewelry fabrication process which includes task such as cutting, grinding, filing and polishing in buffing wheels, sawing and others (Kaspin and Mohamad 2015). Although the amount of dusts released during gold jewelry manufacturing are small and would not be perceived as a major source of pollution, studies have shown that toxic metals at substantial concentrations were evident in blood measured via biological monitoring of workers (Caroli et al, 1998). Furthermore, dust particles can be found on the floor, work station surfaces, pipes and even on the clothing materials. It is possible that goldsmiths working in small gold workshops may have been continually exposed to high heavy metal levels.

Based on the present literature, there have been a limited number of studies on heavy metal exposures among goldsmith who handle the production of jewelries. In Malaysia, there have been no studies as of yet which have measured heavy metal exposure via biological monitoring among gold jewelry workers. There is a need to provide a baseline data of heavy metals exposures among goldsmith in order to compare it to existing studies. This is to ensure workers were not subjected to unacceptable level of heavy metal exposures that have been linked to deleterious health effects such as cancers. As such, this study was performed with the objective to determine the exposure to lead (Pb) and copper (Cu) in hair samples of goldsmiths in Kota Bharu, Kelantan, Malaysia. Pb and Cu are priority pollutants as reported by the USEPA. Moreover, Pb has been classified as carcinogenic, affecting human health either from occupational or environmental exposure (Rim et al. 2013). In addition, Pb and Cu are listed as toxic priority substances by ATSDR in 2013.

### 2. Methodology

This was a cross-sectional study conducted in 2014 among goldsmiths employed in small and medium sized establishments of gold workshop in Kota Bharu, Kelantan. In total, 10 workshops took part in this study. As these workshops were small in size, the number of employees was generally less than five people. All workers in these workshops were generally multi-skilled and performed goldsmithrelated work as such grinding, casting, machining, polishing, and plating of jewelry. Thus, the population of this study consisted of all workers employed in the participating workshops. In total, a number of 50 workers were invited to take part in the study of which 48 of the workers gave their consent for participation.

### 2.1. Questionnaire

Upon receiving the workers' consent to participate in this study, they were given a self-administered Malay version questionnaire that was used to obtain information regarding background information, work history. Next, the workers were briefed regarding the method for hair sample collection. Approximately 100-300 mg of hair samples were collected for each subject from the occipital zone of the head at 1 cm from the scalp using surgical scissors as suggested by Palimi et al. (2013). The collected samples were placed in sealed polyethylene bags pending transport to laboratory. The hair samples were stored in a desiccator in a dark room pending analysis.

#### 2.2. Hair Analysis at Laboratory

Before analysis, the hair samples were prepared and digested using hair analysis method (D'Ilio et al, 2000) at the Bioscience Institute Laboratory, Universiti Putra Malaysia. The steps included the following; the collected hair samples were cut into smaller pieces and then the samples were added with 50 ml deionized water (60°C) and kept for 30 minutes. Next, the hair samples were washed for three times with ultra-pure water and were next dried in a drying oven for 15 minutes at 105°C.

For digestion, approximately 300-400 mg of hair samples in 100 ml beakers were added with 2 ml 69% concentrated nitric acid or HNO<sub>3</sub> and were left to predigest for 30 minutes. The hair samples were then placed in the microwave digester at 80°C for 45 minutes. The digested samples were then diluted to 15 ml liquid using deionized water. At this stage the samples were then was ready for analysis. The final volume obtained was 25 ml. The digested samples were then placed in 50ml Schott bottle, labelled, and were stored at room temperature until analyses. The hair sample were then analysed using Inductively Coupled Plasma – Optical Emission Spectrometry (ICP-OES) Optima 2000 DV (PerkinElmer, Waltham, MS, USA).

### 2.3. Quality Control and Data Analysis

Quality control method was implemented when the hair samples were analysed. The quality controls were performed by replicating the sample analysis for three times. The calibration standards were between zero ppm to 100 ppm. This method is used as to prevent outliers in the result because of possible unseen contamination with the standard solutions. The questionnaire was back-to-back translated from English to Malay. Before the field study was performed, the questionnaire was pre-tested among ten workers in order to identify and amend any ambiguous terms. The questionnaire data were entered into SPSS Version 22 (IBM Corporation, NY, USA). All Pb and Cu levels were expressed in µg/g and averages were calculated using arithmetic and geometric mean (GM) formulas. For statistical analysis, log values of Pb and Cu was used.

## 3. Results

The response rate for this study was 80%. From 48 respondents who took part in this study, 8 of them were either salesperson or account clerk, both of which do not perform the task of producing gold articles, thus they were excluded from the analysis. In total, 40 goldsmiths were included in this study. In terms of socio-demographic distributions, the results in Table 1 showed that among the goldsmiths, there were higher percentages of males when compared to females. More workers had diploma level of education and a larger percentage of them had the income level between RM 1000-2999. More than one-third of the respondents were current smokers.

# **3.1.** Lead (Pb) and Copper (Cu) distribution in hair samples

In terms of Pb exposure, all workers had Pb exposure and the mean (standard deviation; SD) of Pb was 72.0 (107.3)  $\mu$ g/g. On the other hand, the GM of Pb levels in hair samples was 31.9 µg/g. For Cu levels in hair samples, the mean (SD) was 35.4 (22.9)  $\mu g/g$ while the GM was 28.9 µg/g. Female respondents had significantly higher Cu levels compared to male while smoking was associated with lower Cu levels. None of the other socio-demographic factors were associated with the heavy metals distribution. Further analysis performed between the distribution of logged Pb and logged Cu values found that there was a significant positive correlation (r=0.36, p=0.023) between the two metals in the hair samples as presented in the scatterplot in Figure 1 (Pb and Cu distribution was converted to log values before correlation analysis was performed).

This cross-sectional study was conducted among goldsmiths working in small gold workshops in Kota Bharu, Kelantan. The importance of this study lies on the exposure assessment of chronic toxic Pb and Cu concentration in hair samples among a specific type of occupation. As have been reported in ATSDR, the heavy metal levels in the first 2 cm proximal to the scalp would represent heavy metal intake for over two months period (ATSDR, 2004).

#### 3.2. Work Factor and Safety and Health Practices

Table 2 presents the reported work factors and safety and health practices among the goldsmiths. It was found that approximately half of all workers have less than 9 years of experience (45%) and most of them performed different tasks of work including goldsmith work (17.5%), designing (4%) and both task including performing the task of a salesperson. The working hours in a day was mostly 8 hours (92.5%) more than half of them reported overtime work in a week (67.5%) averaging for more than 4 hours (79.3%). The goldsmiths reported additional work during holiday seasons (72.5%) and reported that the major hazard of their work is both exposure to dust and chemicals (88%). Further analysis found that Cu levels were significantly associated with higher overtime duration.

goldsmiths					
Variables	N (%)	GM	<i>t</i> -test	GM	<i>t</i> -test
(n=40)		(Pb)	value	(Cu)	value
		μg/g		μg/g	
Age					
20-29	13 (32.5)	21.90		32.58	
30-39	17 (42.5)	39.14	$0.87^{a}$	28.94	0.45 <sup>a</sup>
>40	10 (25.0)	36.93		24.86	
Gender					
Male	37 (92.5)	32.92	0.87	26.77	2.83**
Female	3 (7.5)	22.0	0.87	76.14	2.05
Ethnicity					
Malay	39 (97.5)	31.04	-	29.16	-
Indian	1 (2.5)	97.36		21.81	
Marital					
status					
Single	11 (22.9)	32.99	0.00	35.92	1.07
Married	29 (77.1)	31.55	0.09	26.67	1.27
Edu. level					
2°	28 (27.5)	30.85	0.00	27.34	0.00
3°	12 (72.5)	34.63	0.26	33.08	0.82
Income					
(RM)	4 (10.0)	23.97		12 00	
<999	30 (75.0)	33.20	$0.11^{a}$	42.98 27.44	0.79 <sup>a</sup>
1000-2999	6 (15.0)	31.86	0.11	27.44	0.79
>3000				29.09	
Height (cm)					
<149	1 (2.5)	14.04		68.79	
150-169	19 (47.5)	37.04	0.39 <sup>a</sup>	30.94	$1.20^{a}$
>170	21 (50.0)	28.92		26.03	
Weight (kg)					
50-69	21 (52.5)	35.82	0.60	29.55	0.20
>70	19 (47.5)	28.14	0.60	28.30	0.20
Current					
smoker					
No	26 (65.0)	34.90	0.60	36.02	3.12**
Yes	14 (35.0)	27.10	0.00	19.29	5.12

 Table 1. Socio-demographics distribution of

\*\* *p* less than 0.001 a: One-way ANOVA F value





**Table 2.** Distribution of Pb and Cu in hair samples

	Mean (SD)	GM	min-max
Pb (µg/g)	72.00 (107.34)	31.94	4.45-402.85
Cu (µg/g)	35.42 (22.97)	28.95	5.55-93.26

Table 3 presents the distribution of safety and health practices among goldsmith and the corresponding Pb and Cu levels. It was found that approximately two-third of the workers reported the use of personal protective equipment (PPE) (77.5%) which mainly consist of respirator (51.6%). The PPE used during working hours in a day was mostly for less than three hours (96.6%) and approximately half of them reported that they were given PPE training (52.5%). Approximately half of the goldsmith reported that they were exposed to hot metal fumes (42.5%) while they are at work. When heavy metal distribution were analyzed further across the safety and health practices, none of the factors were found to be significantly associated lower distribution of Pb and Cu levels.

## 4. Discussion

Chronic occupational exposures to Pb have been with toxicity effects associated such as neurobehavioral decrements (Schwartz et al, 2001) and renal impairments (Kim et al. 1996) in individuals exposed to lead. As reported by the Agency for Toxic Substances and Disease Registry, long-term exposure to Cu has been associated with damage to the liver and kidneys (ATSDR, 2004). A recent study found a significant association between abnormal Cu and manganese levels and increased risk of prostate cancer (Karimi et al, 2012) in a study in Malaysia. The role of Cu in carcinogenic process is linked with its angiogenic activity (Banas et al, 2010). Furthermore, Cu mediates the involvement of cellular proliferation thus it is likely that Cu promote cancer by increasing blood supply for tumour growth.

There is no specific available standard to be referred as guidance to compare the safe reference range for the heavy metal exposure in human hair samples. There are occupational-related legislative standards available in Malaysia as a reference for Pb measurements in biological samples namely blood.

Variables	Ν	GM	t-test	GM	t-test
	(%)	(Pb)	value	(Cu)	value
Work exp.					
(years)					
<9	18 (45.0)	24.80		28.65	
10-20	15 (37.5)	44.80	0.922 <sup>a</sup>	29.77	0.023 <sup>a</sup>
>20	7 (17.5)	29.66		28.00	
Work sect.					
Goldsmith	7 (17.5)	39.76		27.49	
Designer	4 (10.0)	39.57	0.219 <sup>a</sup>	20.04	0.749 <sup>a</sup>
> 1 section	29 (72.5)	29.41		30.84	
Working					
(hrs/day)					
8	37(92.5)	32.63	0.373	29.01	0.063
6	3 (7.5)	24.57		28.28	
Working					
(hrs/week)					
6	40 (100.0)	31.94	-	28.95	-
Overtime					
work (in a					
week)					
No	13 (32.5)	26.41	0.661	34.72	1.203
Yes	27 (67.5)	35.00		39.39	
Overtime					
(hrs)					
<3	6 (20.7)	17.64	1.423	12.59	3.458**
4>	23 (79.3)	39.55		31.83	
Peak					
season					
Celebration	29 (72.5)	37.27		32.90	_
Others	3 (7.5)	10.39	1.58 <sup>a</sup>	16.70	2.287 <sup>a</sup>
More than	8 (20.0)	28.15		22.39	
season					

**Table 3**. Distribution of work history (n=40)

\*\* p less than 0.001 a: One-way ANOVA F value

 Table 4. Characteristics of safety and health practices

across Pb and Cu levels (n=40)					
Variables	N (%)	Pb	t-test	Cu	t-test
		(µg/g)	value	(µg/g)	value
Using PPE					
(n=40)	9 (22.7)	17.64		21.13	
No	31 (77.5)	37.94	2.373*	29.85	1.659
Yes	()				
PPE that has					
been used <sup>€</sup> (n=31)					
(II-31) Respirator	14/21 0			20.04	
Respirator	16 (51.6)	24.30	2.049	30.04	0.527
and glove	15 (48.4)	61.05		33.63	
U					
Use PPE					
during work					
(hrs) (n=29) >4 hours	1 (2 4)	380.3		80.27	
4 hours <3 hours	1 (3.4) 28 (96.6)	380.3	-	30.54	-
PPE training	28 (90.0)	31.23		50.54	
(n=40)					
(II 10) No	19 (47.5)	23.10	1.584	25.67	1.085
Yes	21(52.5)	42.82		32.27	
Exposure to	× ,				
metal fumes					
(n=40)					
No	23 (57.5)	30.73	0.225	27.24	0.664
Yes	17 (42.5)	33.66		31.42	
* <i>n</i> less than 0.05 $^{\circ}$ answers 'Yes' on previous					

\* p less than 0.05 <sup>c</sup> answers 'Yes' on previous

question

The Use and Standard of Exposure of Chemicals Hazardous to Health Regulations, 2000 under the Occupational Safety and Health Act, 1994 and the Factories and Machinery (Lead) Regulations 1984 have specified the requirement for medical surveillance among workers who are exposed to Pb (Rampal and Jemoin, 2006). No standards for Pb are available for hair samples except for blood lead levels. On the other hand, there are no occupational biological monitoring standards available for Cu.

In comparing the Pb and Cu distributions with a previous study performed in Italy among 75 goldsmiths, this study reported extremely high Pb exposure (0.66  $\mu$ g/g) and Cu levels (13.27  $\mu$ g/g) (D'Ilio et al, 2000). The difference between the study done by D'Ilio et al. (2000) in Italy and this study was the focus on the production of jewelries in which the complete working cycle from melting to casting was carried out by a single craftsman (D'Ilio et al, 2000). Whereas in this study the workers multi-tasked and performed work such as designing gold jewelry to performing tasks of a goldsmith. Despite this fact, the Pb and Cu levels remain high, evident of the need for safety and health measures to be employed.

— One study of heavy metal distribution in hair samples among selected groups of employees was reported by Shan and Ikram in Pakistan (2012). The study reported that the levels of Pb among forty jewelers were lower than the present study, being between 10.5 to 20.9  $\mu$ g/g. The levels for Cu was higher compared to the present study, with levels varying between 38.6 to 130.8  $\mu$ g/g. Interestingly, the study by Shan and Ikram (2012) showed that the Pb levels were only comparable to the group of industrial workers rather than the jewelers. This shows that the goldsmith in the present study is exposed to a substantial exposure to Pb as evident from the hair biomonitoring.

Compared to another study performed locally in Malaysia, Pb and Cu levels in human hair from male subjects working as sanitation handlers was found within the range of 38.0 to 39.0 µg/g and 684.0 to 962.0 µg/g respectively (Khudzri et al, 2013). The results from Khudzri et al. (2013) clearly indicated that Pb levels in hair samples of goldsmith in this study were almost two-fold higher compared to sanitation handlers. These evidences further strengthens the fact that the goldsmith in the present \_study have high toxic metal exposures and this clearly spells out the need to have control measures put in place to reduce the existing exposures. However the levels of Cu in the present study is much lower than the extraordinarily high levels reported among sanitation workers. When compared to Cu levels among forty fisherman in India, the levels were within the range 6.1 to 35.3  $\mu$ g/g, similar to the range found in this study (Subramanian and Ramachandran, 2003).

This study has revealed that Pb and Cu levels in hair samples does not fluctuate significantly across age, duration of work, ethnicity, education or income level and height or weight. Female goldsmiths had higher Cu levels but the average was for a small number of respondents (N for female=3) to be meaningful. On the other hand, current smoking was associated with lower Cu levels instead of the contrary. It may likely be that the non-significant findings was due to small number of sample size observed in this study which does not allow for further statistical analysis to be performed.

Overtime work was associated with higher Cu levels while for Pb, the use of PPE was not found to reduce the levels in hair samples. None of the other work factors and safety and health practice was linked with the Pb and Cu distributions. It is a common in Kelantan that small gold workshops which usually have limited number of skilled goldsmiths shared goldsmiths with other workshops at required times. Thus, the exposures to heavy metal related to occupational task in this study may not be localized from only one source in the sampled workshops and thus rendering the search for associations with work factors and safety and health practice difficult.

When reported safety and health practice was taken into account, it was clear that not all workers were given effective training on control of exposure methods. A previous study among goldsmiths has shown that lack of proper knowledge about workplace environment leaves the workers highly vulnerable for work-related injuries and accident (Sahu et. al, 2013). In the present study, not all workshops applied engineering controls such as Local Exhaust Ventilation (LEV) system and when LEV is available, it is only used at certain workstation. Furthermore, the LEV system was not applied during all period when work was performed but is used when high extractions of dust were required only. Moreover, a number of managers reported that no maintenance of the LEV system was provided. For workshops that are small in size, the implementation of LEV system was not feasible.

This study has a few limitations to be considered. This study did not consider environmental exposures that may lead to heavy metal exposures and dietary habits as one of the heavy metal contributions. This study also did not measure the airborne contaminations of Pb and Cu exposed to each of the worker. It may be likely that measurements of airborne contaminations of Pb and Cu will give a better picture of where the highest exposure takes place in order to better understand about the factors affecting the Pb and Cu exposure. Notwithstanding these limitations, the strength of this study includes the selection of hair as the biological sample of interest. Keratinous tissue such as hair retained heavy metal deposited over a length of time, detached from the metabolic process after it is formed and is suitable to represent long term exposure (Ab Razak et al, 2015; Morais et al, 2012). As such hair samples could represent a more integrated measure of exposure over a long period. The measurement of biological samples in this study was performed using ICP-OES, the most sensitive elemental spectrometric technique with lower detection limits which enables the detection of low levels of heavy metal concentrations (Morais et al, 2012) as compared to atomic fluorescence spectrometry or atomic absorption spectrometry.

The importance of this study includes the fact that there is a gap in the practice of occupational safety and health for this subgroups of workers. This study further provides evidence on the level of occupational exposure among a group of workers in a workplace with limited access to researchers as compared to other common industries. Such quantification of levels may be of interest to occupational health professionals for future regulation purposes, or to assist in diagnosis of occupational disease in the future.

# 5.0 Conclusion

In conclusion, the mean (SD) and GM of heavy metals in hair samples of goldsmith were 72.0 (107.3)  $\mu$ g/g and 31.9  $\mu$ g/g respectively for Pb while for Cu the levels were 35.4 (22.9)  $\mu$ g/g and 28.9  $\mu$ g/g respectively. The levels reported in this study exceed the levels reported in other recent studies. Longer overtime work was associated with higher heavy metal levels. The reported safety and health practice was poor and the usage of PPE was not linked to lower heavy metal exposures. Thus, some of the measures that can be adopted are the use of suitable LEV system at work sections that produce dust and fumes. This study has showed that there is a need for safety and health training to be given to goldsmith to increase their knowledge and practice in safety and health and to protect them from the exposure to toxic heavy metals. New workers needs comprehensive trainings before they start work and continuous education program in order to encourage safety precaution during work. This study has also shown that there is a need for this specific group of workers to be continuously monitored for exposure assessment in order to lower their occupational-related heavy metal exposures. Exposure measurements are essential for the protection of high risk populations and subgroups.

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# **ETHICAL ISSUES**

The Ethical Committee for Research involving Human Subjects of Universiti Putra Malaysia approved this study. The respondents were given and explanation about the entire of process measurement and evaluation along this research. All the information and identity used in this study remains confidential.

# **CONFLICT OF INTEREST**

The authors declare that there is no conflict of interest in the publication of this research.

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