VOL. 9, NO. 12, DECEMBER 2014 ISSN 1819-6608
ARPN Journal of Engineering and Applied Sciences

www.arpnjournals.com

AN ASSESSMENT OF SELECTED HEAVY METAL CONCENTRATIONS (Pb, Cu, Cr, Cd, Ni, Zn) IN UNIVERSITY CAMPUS LOCATED IN INDUSTRIAL AREA

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ABSTRACT

An investigation was conducted to examine the variation trend of heavy metal concentration and distribution in ambient air in university campus located in the industrial area. The aim of this study was to measure the concentration of heavy metals of concern (lead (Pb), copper (Cu), chromium (Cr), cadmium (Cd), nickel (Ni) and zinc (Zn) composition). The heavy metals were chosen based on the types of industry. Three sampling points were set up according to the distance from the industrial area. The sampling points were located at the Convocation Hall (3, 000m), Tun Dr. Ismail Residential College (390 m), and the Material Laboratory (10 m) in the Universiti Tun Hussein Onn Malaysia (UTHM) campus, and all of the locations were in the Parit Raja industrial area. Heavy metals particulates were sampled from the E-Sampler Particulate Matter (PM_{10}) Collector (Met One Instrument, Inc) in 24 h period for samplings of October 2013 to Jun 2014. Determination of heavy metals concentrations was conducted using two instruments; Atomic Absorption Spectrometer (AAS) (Perkin Elmer Elan 900) and Inductively Coupled Plasma-Mass Spectrometry (ICP-MS) (Perkin Elmer AAnalyst 800). The analysis was performed after the filter paper was digested using aqua regia method. The relationship between metrological factor and concentration of heavy metals were clarified using Pearson's correlation coefficient. The data were compared to the air quality standards by the World Health Organization Health (WHO), the Ambient Air Quality Criteria Act 1994 (AAQC), the National Environment Protection Council (NEPS), Texas Commission on Environmental Quality (TCEQ), and also the Environmental Quality Act 1974 (EQA). It was found that the concentrations of Cr, Cd, and Ni exceeded the standard concentrations which were 0.0100 $\mu g/m^3$, 0.0050 $\mu g/m^3$ and 0.0150 $\mu g/m^3$, respectively. In contrast, Pb, Cu, and Zn in ambient air had low concentration compared to the standard, which were 0.500 $\mu g/m^3$, 1.0000 $\mu g/m^3$ and 1000 $\mu g/m^3$, respectively. Generally the nearest distance of sampling site to the industrial area was indicated high levels of heavy metals. This finding is significant in contributing some knowledge of the heavy metals concentration in ambient air to communities surrounded by an industrial district. It identifies research needs and suggests possible approaches to addressing outstanding questions.

Keywords: heavy metals, air quality, ICP-MS, AAS, industrial area.

INTRODUCTION

Intensive anthropogenic activities are caused by the high density in population, where population will produce a large number of heavy metal sources that have considerable influence on human health [1-2]. Some of the major sources that emit heavy metals into ambient air and its surroundings are domestic, transportation, and industrial wastes. Several researchers have sought the assessment of heavy metals in ambient air as affected from various activities. A research in Coimbatore city, India was found that the level of heavy metal was significantly high and extensive research is needed to relate the possible factors and its consequences [3]. The research conducted in Shanghai Nanjing Expressway and industrial area in Balakong, Malaysia had proven that the main pollutants were Zn and Pb [4-5]. In the Howard University, Washington, the concentrations of Pb were 137 ng/m^3, which might be contributed from the vehicles in the university itself [6]. Also, in Greece, research indicates that the concentrations of Cd, Pb and Ni during summer were 1.06 $\mu g/m^3$, 12.23 $\mu g/m^3$ and 13.3 $\mu g/m^3$ respectively [7].

As one of the Southeast Asian nations, Malaysia is considered as new in the industrialized sector [8-9].

Even though the development of industries in Malaysia gives a great benefit to the Malaysian economy, but it is also a major source of pollution towards the environment. Pb, Cu, Cr, Cd and Ni are several general types of pollutants that are being emitted by factories. A study was conducted by Sulaiman (2006) in Balakong in order to determine the concentration and composition of heavy metals of PM_{10} for indoor and outdoor air quality. The outdoor air quality for two factories was differentiated according to the smoke activities. For Factory 1 that produced smoke, 91.92 mg/m^3 and 0.30 mg/m^3 of Zn and Cr were detected respectively. Meanwhile, for Factory 2 that did not produce smoke, there was no Zn and Cr undetected in the outdoor air. Therefore, industrial district is indeed responsible for the phenomena of air pollution that occur nowadays.

It is very important to study the concentration of heavy metals in ambient air in order to determine the level of air quality. As Universiti Tun Hussein Onn Malaysia (UTHM) campus is located nearby an industrial area, it has raised the concern of air quality towards students and

VOL. 9, NO. 12, DECEMBER 2014 ISSN 1819-6608
ARPN Journal of Engineering and Applied Sciences

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university staff. Consequently, this research is conducted in order to determine the level of air pollution that occurs in ambient air of the UTHM campus. The concentration of heavy metals acts as an indicator in assessing the level of ambient air in UTHM campus. Three objectives were considered in this research, which were to determine the variation of heavy metals concentrations at each station, to identify the distribution of heavy metals during wet and dry days, and to measure the rate of heavy metals concentrations with the standard allowed by the standard concentration of ambient air.

Previous studies on heavy metals in ambient air

Due to high awareness about the quality of ambient air, a lot of research have been undertaken to prove the existence of heavy metals in air. Previous research has shown that issues of concentration of heavy metals in the air are not only worrying in Malaysia, but also to the global community. Previous studies on heavy metals worldwide are shown in Table-1. From Table-1, most of the research focus on industrial and traffic flow as the targeted sources. Their researches have proven that heavy metals do exist.

Case study description: parit raja industrial area

The types of industries that are located in the Parit Raja industrial area are timber, semiconductor and packaging industries. Effluents from industries contain an appreciable amount of metallic cations such as zinc (Zn), copper (Cu), iron (Fe), manganese (Mn), lead (Pb), nickel (Ni) and cadmium (Cd). Fiber board is the main product that is produced by one of the factories in the Parit Raja industrial area. Use of heavy metals can be perceived in the treatments of the timber. Zn and Cu are used in chemical solution, where timbers absorb this solution well [11]. This chemical solution can prevent woods from being attacked by fungus and insects, but the solution has a pungent smell. Another method to preserve the timber is by using water preservation. This current timber treatment designed to prevent wood decay is based on copper-chromium-arsenic (CCA) pesticides impregnated into wood to give total concentrations between 100 and 4,000 mg/kg for each element [12]. It is stated that this CCA method uses a high concentration of heavy metals in order to preserve the wood.

Table-1. Previous study of heavy metals worldwide.

Research	Location	Targeted sources	Detected heavy metals						Unit
			Cd	Pb	Cr	Ni	Cu	Zn	
Vassilakos (2007)	Greece	Wood burning and smoke from vehicle	1.06 √	24.7 √	-	13.3 √	-	-	ng/m³
Melaku (2008)	Howard University, Washington	Heavy traffic flow	30.0 **x**	137 √	112 **x**	-	-	-	ng/m³
Vijaynand (2008)	Coimbatore, India	Various sectors	ND	0.58 **x**	0.88 **x**	0.22 **x**	0.92 √	24.96 √	μg/m³
Zhao (2012)	Shanghai-Nanjing Expressway	Smoke from vehiclesthat passed through the expressway	0.16 **x**	0.35 **x**	0.37 **x**	0.32 **x**	-	165.8 **x**	mg/m³
Sulaiman (2006)	Industrial Area, Balakong	Emission from factories	-	-	0.30 **x**	-	-	91.92 √	mg/m³

[Symbols]
'√' = complied with the standard of heavy metals listed in Table-2
'x' = did not comply with the standard of heavy metals listed in Table-2

In the Parit Raja industrial area, fiberboard, corrugated paperboard, and heavy duty board are the example of products that are being produced by certain factories. For the production of fiberboard, the raw materials used are saw mills and plywood plants. Sawmill is a wood waste that may also be applicable as a raw material for the medium density fiberboard industry [13]. Therefore, it can be said that there are possibilities of the saw mills that are being used is a waste of timber that has undergoes preservation using heavy metals.

Semiconductor industry uses many chemical substances to produce semiconductors including heavy metals such as Cd, Cu and Pb [14, 15. In order to produce semiconductor plate, the processing steps involved include preparation of wafer, front-end line-of-line (FEOL) processing, back-end-of-line processing (BEOL), test and packaging. The use of heavy metals in the production of semiconductor can be linked with the production of wafer. A wafer is a thin slice of semiconductor material that is used in the fabrication of integrated circuits [16]. Semiconductor materials act as an insulator, and their application in the device is similar to a transistor. Cadmium telluride (CdTe) is one of examples used in the

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semiconductor industries, where CdTe is used to produce thin film solar cells [17]. Most of the packaging products that are produced by the industrial area in Parit Raja are made up of paper. Also, the heavy metals used in packaging product are mostly chemicals that are used to add up color. Packaging, dye substances and ink pigments, and chemical manufacturing industries always useheavy metals such as Pb, Cu, Cd, Ni and Cr. In order to reduce thecost, heavy metal like Pb is used in ink manufacturing by blending with heavy metals pigment [18]. For dye substances, heavy metals are needed to produce color such as orange (Cd), black (Ni), and green (Cr).

Figure-1. Location of the university adjacent to the industrial Area (retrieved on 21st December, 2013, googlemap.com)

Standard concentration of heavy metals

The data of the standards for heavy metals in ambient air from different regulations are combined and presented in Table-2, and the data can be used as a standard for comparison purpose with the concentration of heavy metals in the UTHM area. This is due to unavailable standard of heavy metals in Malaysia.

Table-2. Heavy metals with their standard concentrations in ambient air.

Heavy metal	Standard concentration (μg/m³)	References
Cd	0.005	WHO, AAQC
Pb	0.500	NEPC, AAQC
Cr	0.010	TCEQ
Ni	0.015	TCEQ
Cu	1.000	TCEQ
Zn	1000	EQA

References such as the Ambient Air Quality Criteria Act 1994 (AAQC), the World Organization Health (WHO), Texas Commission on Environmental Quality (TCEQ), the National Environmental Protection Council (NEPS) and the Environmental Quality Act (EQA) 1974 have been referred to. All the standards are listed in Table-2.

MATERIAL AND METHODS

The total number of ambient samples for this study was 29. In order to distinguish samples, the sample was identified as Sample 1 for the first day, Sample 2 for the second day and others. For each station, ambient samples were collected three days in November 2013, which were Sample 1, Sample 2, and Sample 3. Meanwhile, seven other samples were collected during January to Jun 2014.

Site description and sampling

For this study, the level of UTHM ambient air was monitored at three points, which include background, residential and public area. The Convocation Hall was the first sampling site, and it acted as the background since it was located the farthest from the factory. After that, the chosen sampling site was at the Tun Dr. Ismail Residential (TDI) College and the Material Laboratory. Material Laboratory was selected in this study due to its location beside the factory and used as a public area. Identically, TDI was chosen based on its location nearby the factory, and it was also the most packed place in terms of population. By using E-Sampler Particulate Matter (PM10) Collector (Met One Instrument, Inc), the equipment was setup at the chosen locations. Filter paper acted as a medium to collect heavy metals in air. The filter paper then was changed every 24 h**,** which is in accordance to the sampling duration stated by Vassilakos (2007).

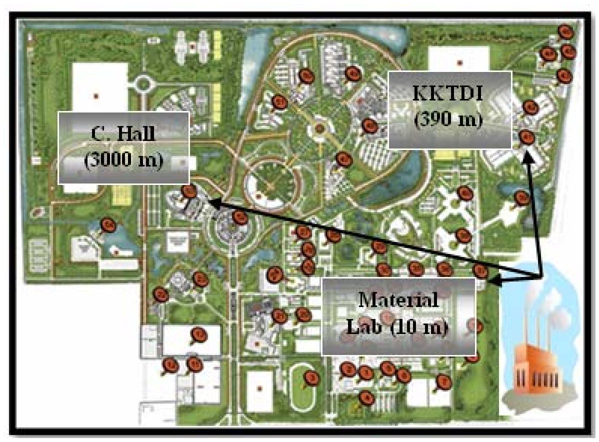

Figure-2. The distance of sampling locations from the nearest factory (Pelan Induk UTHM, 2013).

Analysis procedure

AAS (Perkin Elmer Elan 900) and ICP-MS (PerkinElmer AAnalyst 800) were used to analyze the detected heavy metals. One of the limitations for these instruments is that the instruments can only analyze a liquid sample. The trapped particulate matters were in the

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form of solid. Therefore, digestion of the samples was done to obtain the liquid form of the particulate matters. There are two methods to digest a sample from the filter paper, which are by using microwave or acid extraction [11]. In this study, the aqua regia digestion method was used, which is a type of acid extraction. The whole mixture was heated on the hotplate for 2h. According to the International Organization for Standardization ISO 11466, samples should be digested at 130ºC for 2 h under reflux condition [6]. This process was done in a fume chamber. The digested sample needs to be preserved so that it can be kept for a long period. After the preservation step has been completed, the chosen acid with its respective concentration should be added until the sample has a pH less than 2. However, for this study, the addition of acid was unnecessary because the sample itself had a pH of less than 2. In order to get the desired concentration, the sample was diluted in order to reduce the concentration of the sample. As the sample was digested by acid, the concentration of the sample was in accordance with the concentration of acid. The dilution was performed as the ICP-MS and AAS have rough detection limit [11].

ANALYSIS AND DISCUSSIONS

Analysis on the variation of heavy metals

The daily variation of heavy metals is important in order to determine the correlation of surrounding condition and meteorology factors with heavy metals. By using Sigmastat (12.0), Pearson's correlation coefficient was used to associate heavy metals and metrological factors such as humidity and wind speed. In this research, Cu and Cd showed a vibrant variation at all sampling stations. Figure-3 presents the variation of Cu at each station. Sample 4 presented the maximum daily concentration of Cu of 0.0359 μg/m³, and it was detected at the Convocation Hall. This was followed by the TDI College with maximum daily concentration of 0.032 μg/m³. Meanwhile, the highest concentration of Cu at the Material Laboratory was detected as 0.0276 μg/m³.

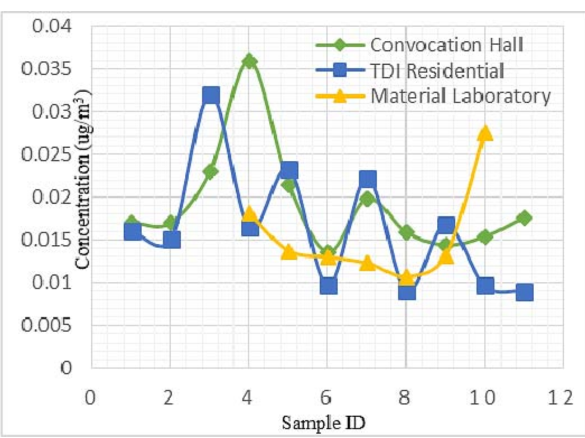

Figure-3. Variation of Cu concentrations at sampling stations in UTHM campus.

Figure-3 shows the variation of concentrations of Cu in the Convocation Hall and TDI College, which were mostly higher than the concentrations of Cu detected at the Material Laboratory. At the Material Laboratory, the statistical analysis indicated a high positive coefficient of Cu with wind speed ($r = 0.75$) and humidity ($r = 0.522$). The correlation coefficient between these two measurements suggested that the high concentration of Cu was affected by high relative humidity and high wind speed at the sampling area. The existence of Cu at the TDI College and the Convocation Hall might be associated with the traffic volume located nearby the station. High number of parking lots near to both stations also increased the concentration of Cu as a result of uncontrolled emissions, as well as Cu particles resulting from the friction of the tires of motor vehicles with road.When tire tread is abraded against the road surface, the tire tread debris will assimilate heavy metals [19]. When the traffic volume is relatively high, the exposure of people to the traffic related concentrations is also significant. Increase in traffic volume can also be associated with an increase in the number of registered vehicles in the UTHM campus. In 2012, the total registered vehicles were 10, 232. There was a slight increase of the total registered vehicles in 2013, with a total of 10, 509 vehicles.

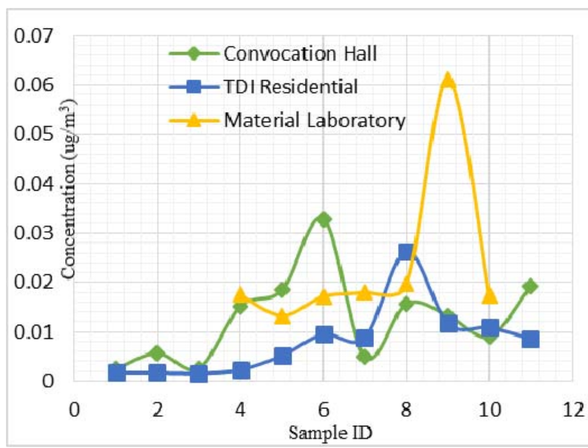

Figure-4. Variation of Cd concentrations at sampling stations in UTHM campus.

Figure-4 presents the variation of Cd at each station. The concentrations of Cd ranged from 0.01327 μg/m³ to 0.0611 μg/m³ for the Material Laboratory, 0.0025 μg/m³ to 0.0329 μg/m³ for the Convocation Hall, and 0.0016 μg/m³ to 0.0263 μg/m³ for the TDI College. The highest single concentration for Cd determined at the Material Laboratory was 0.0611 μg/m³, and followed by the Convocation Hall with daily concentration of 0.00329 μg/m³, which was detected on the sixth day of the sampling. For TDI Residential, 0.0263 μg/m³ of Cd was detected. Vibrant variation of Cd was present at the Convocation Hall. The positive correlation coefficient between these three measurements suggested that the high concentration of Cd was affected by high relative humidity

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(r = 0.949) and high wind speed (r = 0.698) at the sampling area. Regression analysis showed a positive relationship between both metrological factor and Cd. Cd might travel from the industrial area by wind. The contaminations of Cd at the Material Laboratory might be related to the production of semiconductor from the industrial area that was located beside the UTHM Campus. Cd is used in the production of wafer, which is one of the semiconductor components. Cadmium tellurides (CdTe) are the most common photovoltaic (PV) materials used in the mass production of thin film modules at present [17]. Meanwhile, the contaminants of Cd at the Convocation Hall may be caused by traffic that passed by near the road because cadmium may come from lubricating motor oil, tires and galvanized parts of vehicles [20].

Distribution analysis for concentration of heavy metals during dry and wet days

Table-3 demonstrates the distribution of heavy metals during dry and wet days from the whole sample. Unequal distribution of heavy metals can be seen, where most of the heavy metal showed a high concentration during dry days compared to the wet days. For the Convocation Hall, Tun Dr. Ismail Residential and the Material Laboratory, Zn was detected to the maximum concentration on a dry day, which was 3.3800 g/m^3, 3.9400 g/m^3, and 7.3900 g/m^3 respectively. It was found that during dry days, the air quality for three stations was worse than wet days. However, at the Convocation Hall, the average concentration for the other five heavy metals were relatively high during dry days compared to wet days, except for Cr. During wet days, the average concentration of Cr was 0.161 μg/m³, which was higher than 0.0703 μg/m³ on dry days at the Convocation Hall. Similar trends were also detected for minimum, median and average concentration of Cr at the Convocation Hall, where the concentration was high during wet days. Meteorological factors might contribute to these trends. Small amount of precipitated rain indicated a 40% of humidity at the Convocation Hall, and it was considered as low humidity. Low level of humidity increases the lifetime of particulate matter in the ambient air, thereby resulting in higher concentration of PM10 in the atmosphere (Dey, 201). This wasproven by thenegative correlation of r = - 0.326 between Cr and humidity. Cr concentration wasobserved to exhibit peak concentration value, which was coincident with the lower relative humidity.

Table-3. The distribution of heavy metals concentrations during dry and wet days.

		Pb		Cr		Ni		Cu		Cd		Zn	
		Dry	Wet	Dry	Wet	Dry	Wet	Dry	Wet	Dry	Wet	Dry	Wet
Stn 1	Max	0.0881	0.0200	0.1068	0.2650	0.0847	0.0175	0.0359	0.017	0.0329	0.0057	3.3800	2.3000
	Min	0.0220	0.0170	0.0421	0.0570	0.0121	0.0172	0.0136	0.017	0.0026	0.0025	2.2480	2.2700
	Median	0.0484	0.0185	0.0652	0.1610	0.0200	0.0174	0.0176	0.0170	0.0152	0.0041	2.5800	2.2850
	Average	0.0470	0.0185	0.0703	0.1610	0.0310	0.0174	0.0200	0.0170	0.0146	0.0041	2.6353	2.2850
Stn 2	Max	0.1890	0.1040	0.0610	0.0490	0.0246	0.016	0.0320	0.0160	0.0263	0.0109	3.9400	2.8880
	Min	0.0240	0.0260	0.0322	0.0387	0.0150	0.015	0.0090	0.0096	0.0016	0.0017	2.4000	2.2600
	median	0.0793	0.065	0.0493	0.0502	0.0172	0.0163	0.0164	0.0125	0.0086	0.0052	2.9200	3.1000
	Average	0.1014	0.0650	0.0489	0.0439	0.0181	0.0155	0.0171	0.0128	0.0084	0.0063	2.9433	2.5740
Stn 3	Max	0.4287	0.1815	0.9827	0.2352	0.0385	0.0265	0.0276	0.0137	0.0197	0.0611	7.3900	5.4400
	Min	0.0989	0.1419	0.0746	0.0487	0.0261	0.0229	0.0107	0.0123	0.0172	0.0132	5.5250	4.1000
	Median	0.1235	0.1502	0.1835	0.0703	0.0296	0.0254	0.0181	0.0132	0.0174	0.0176	5.9900	4.8650
	Average	0.2171	0.1560	0.4136	0.1061	0.0314	0.0254	0.0188	0.0131	0.0181	0.0274	6.3017	4.8175

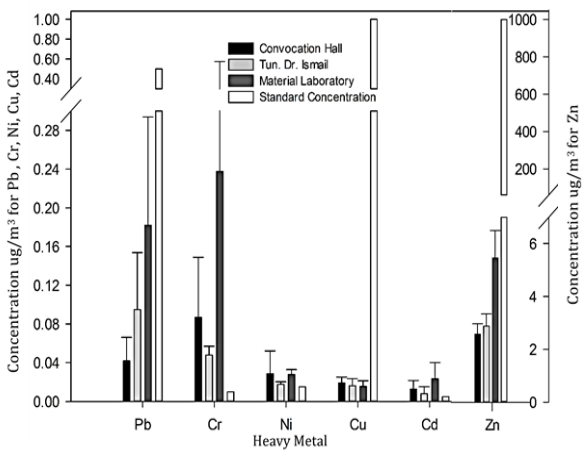

Figure-5. Comparison between concentrations of heavy metals at each station with standard concentration.

From Figure-5, it can be observed that the Material Laboratory had high concentrations for most of the heavy metals compared to the Convocation Hall and Tun Dr. Ismail Residential. The highest concentration of heavy metal detected was Zn at the Material Laboratory, with theconcentration of 5.4535 ± 1.0464 μg/m³. Eventhough the concentration of Zn was high, it was still in the range allowed by the Environmental Quality Act 1974 (EQA), which is 1,000 μg/m³. Pb was detected as the second most concentrated heavy metals. According to the Ambient Air Quality Criteria Act 1994 (AAQC) and the National Environment Protection Council (NEPS), the allowable concentration of Pb in ambient air was 0.5 μg/m³, which was three times higher than the mean concentration that was detected in the Material Laboratory, 0.1821 ± 0.1118 μg/m³. Besides Zn and Pb, a huge difference between the value of mean and standard deviation was noticed for Cr, which was 0.2379 ± 0.3358 μg/m³. By comparing the concentration of Cr with the allowable concentration by the Texas Commission on Environmental Quality (TCEQ), the detected Cr at the Material Laboratory did not comply with the standard concentration (i.e. 0.01 ug/m³). A huge difference of standard deviation and mean concentration might due to the long heavy rain that occurred while conducting the sampling process. This will indirectly cause variation of concentrations of Cr. The data indicated that Cr, Ni, and Cd did not comply with the allowable standards of heavy metals in air. Finally, the occurrence of heavy metals in UTHM ambient air might be ranked as Zn> Pb> Cr> Ni > Cu> Cd.

CONCLUSIONS

It can be concluded from the study that the highest level of heavy metals concentration was detected at the Material Laboratory. High concentration of heavy metals at the Material Laboratory showed that the distance between the laboratory and the industrial area indeed affected the level of heavy metals. From the analysis, it was shown that the daily variation of heavy metals was affected by the surrounding conditions and meteorology factors. Cd concentration was observed to exhibit a peak concentration value that was coincident with the observed humidity (r = 0.949) and wind speed (r = 0.698). Vibrant variation was found for Cu and Cd at all sampling stations. The distribution of heavy metals at the sampling stations in the UTHM campus indicated unequal distribution of heavy metals for dry and wet days. However, all three stations indicated a high concentration in heavy metals during dry days except for Cr at the Convocation Hall. Drizzle and low humidity had contributed to 0.0703 μg/m³ Cr concentrations during dry days and 0.1610 μg/m³ concentration during wet days. From the analysis, it was found that the concentrations of selected heavy metals were high, and some heavy metals such as Cr, Cd, and Ni exceeded the standard concentration. In contrast to that result, Pb, Cu, and Zn detected in UTHM ambient air had low concentration compared to the standard.

ACKNOWLEDGEMENT

The authors are grateful for the financial support provided through the UTHM Contract Grant Vot C006 as part of university's air quality monitoring work. We also appreciate the contributions made by the technicians in Environmental Engineering Lab and also Science officer of Analytical Lab, and academics in Faculty of Civil and Environmental Engineering to the success of this project. We would not have succeeded without their expertise.

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