Ambient air mercury species (TGM, RGM, GEM) concentration study during mid-winter~mid-spring season at a traffic site

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***Abstract*— *The main purpose of this study was to monitor ambient air mercury species (TGM, RGM, GEM) of Hung-Kuang traffic area at sampling site during mid-winter～mid-spring season. In addition, four-stage gold amalgamation and denuder samplers were used to collect the ambient air mercury species concentrations, respectively. Moreover, cold-vapor atomic fluorescence spectrometry (CVAFS) was directly used to detect the mercury TGM and RGM concentrations in this study.*

The result indicated that: 1). The average ratio for RGM concentrations out of the total gaseous mercury concentration was about 1.65% at the traffic sampling site. 2) The average TGM concentration values obtained in this study were 6630 (pg/m^3) which is second to China average 9800 (pg/m^3) when compared with the other countries during year 2010～2013. In addition, Canada showed the average lowest RGM concentrations when Compared the other world areas during year of 2014～2015. The average RGM concentrations ratios for this study (average 91.8 pg/m3) to Canada were about 73.4 during years of 2014～2015. 3) The results indicated that the correlation coefficients among total gaseous mercury (TGM), reaction gaseous mercury (RGM), gaseous element mercury GEM and meteorological conditions such as temperature and wind speed were low correlated related during mid-winter～mid-spring season at this traffic sampling site. 4) Statistical method (Mann-Whitney U statistical) results revealed that there were significant differences in the mean concentrations values for the ambient air TGM, RGM and TGM were the Hung-Kuang sampling periods at this traffic sampling site

Keywords*— *denuder, four-stage gold amalgamation, gaseous element mercury, reactive gaseous mercury, total gaseous mercury.

I. INTRODUCTION

Mercury (Hg) is a trace element distributed throughout the earth's atmosphere, biosphere, and geosphere [1, 2].

Atmospheric Hg consists of three different physical and chemical forms, including gaseous elemental Hg (GEM), reactive gaseous Hg (RGM), and particulate Hg (Hg_p) [3].Atmospheric mercury speciation is very important in the determination of the mercury atmospheric residence time. Atmospheric mercury species can be transformed back and forth between two oxidation states (elemental and divalent) [4] and elemental mercury is believed to have a longer residence time of about 1 year [5] compared to the divalent mercury (several hours to several weeks).

Recent studies reported RGM may represent 1–3% of total gaseous Hg (TGM) at rural continental sites [6,7, 8]. Because RGM is highly reactive and rapidly scavenged by moist particles and surfaces, it can fall off quickly with distance from their primary sources (e.g., incinerators, non-metal smelters and power plants) [9].RGM concentrations are likely to be highly variable and related to point sources, meteorological conditions and oxidant levels in the air [10].

The dominant form of mercury in the atmosphere is identified as a gaseous elemental mercury (GEM: e.g., 98%) that is characterized by high stability, low solubility, high volatility, etc. [11]. Gaseous element mercury (GEM) collection and analysis is quite not easy. Including some potential oxidants include HO, O_3, HO_2, N_2O_5, O (^{1}D and ^{3}P), NO_2, Cl_2, and Br_2[12, 13].

Mercury in ocean waters is present as elemental mercury (Hg0), monomethyl mercury (MeHg), dimethyl mercury (Me_2Hg), aqueous divalent mercury (Hg II), colloidal mercury, and particulate mercury [14]. Among Hg(p) is associated with airborne particles, such as dust, soot, sea-salt aerosols, ice crystal or is likely produced by adsorption of RGM species (eg, $HgCl_2$) onto atmospheric particles [15].

All three forms are released by anthropogenic sources, primarily combustion processes, as well as by a variety of natural sources and processes. Natural sources include crustal degassing, volcanoes, a component of the reemitted mercury from soils and aquatic surfaces, weathering processes of the Earth's crust and some forest fires [16]. On a global scale, the

dominant component of the mercury released from terrestrial and oceanic systems is previously deposited anthropogenic mercury rather than geogenic sources [17]. Contributions from natural sources and processes vary geographically and over time depending on a number of factors including meteorological conditions, the presence of volcanic or geothermal activities, the presence of Hg bearing minerals such as cinnabar, the magnitude of exchange processes between waters and the atmosphere, the re-emission of previously deposited Hg from top soils and plants, and also the occurrence of forest fires [18, 19, 20]. The main purpose of this study is to 1). Monitor ambient air TGM, RGM and GEM concentrations at Hung-Kuang traffic area during mid-winter～mid-spring seasons of Dec. 2015 to Mar. 2016. 2). Compare TGM and RGM concentrations at the Hung-Kuang traffic sampling site with other studies. 3). Find the concentration correlation for TGM, RGM and GEM to meteorological conditions (temperature, wind speed and humidity) at these characteristic sampling sites. 4). Apply appropriate statistical method (Mann-Whitney U) in the testing the mean TGM, RGM, GEM concentration differences during mid-winter～mid-spring season at this traffic sampling site.

II. METHODOLOGY

2.1 Sampling site

Figure 1 display the geographical location at Hung-Kuang University traffic area (24°12'60.0"N 120°35'03.2"E) in central Taiwan. All the samples were collected for 24 hrs. The sampling site is located in Sha-lu, Taichung, Taiwan. Ambient particle concentrations were sampled on the roof of the Medical and Industrial Building at Hung-Kuang University, an eight-story building (22 m height) at the top of Da Du Mountain. There were 6400 vehicles pass by during the day time working hours at this Taiwan Formosa expressed 3 high way. And there were 12000vehicles pass by during the night time working hours at this Taiwan Formosa expressed 3 high way. Basically, it is a traffic characteristic sampling site. Taichung Science Park which is about 9.5 km on the east of sampling site. There were about 298 factories and plants inside the Taichung Science Park. Taichung Harbor Related Industrial Zone and Taichung Industrial Park were located on the southwest, east sides of this traffic sampling site, respectively. Taichung Thermal Power Plant (TTPP) was just on the west side of 13 km away from this traffic site.

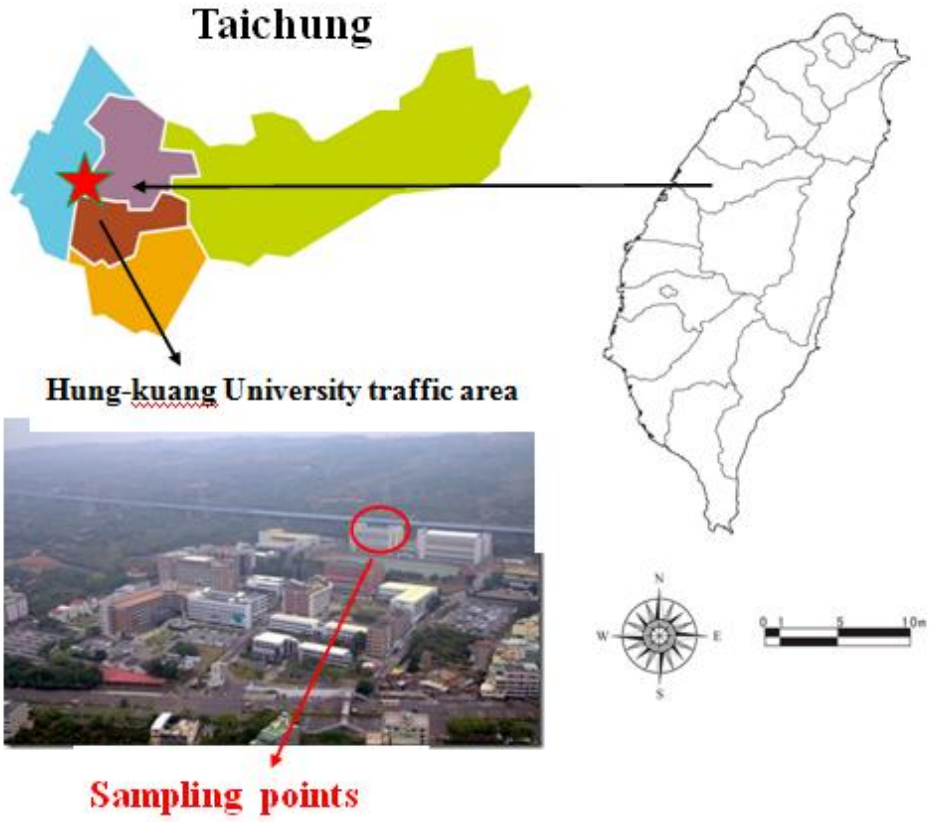

FIGURE 1 CHARACTERISTICS SAMPLING SITE TRAFFIC AT HUNG-KUANG UNIVERSITY TRAFFIC AREA, CENTRAL TAIWAN.

2.2 Four-stage gold amalgamation and denuder

Figure 2-1 displayed four-stage gold amalgamation sampling device. It was used to collect total gaseous mercury (TGM) concentration and was composed of from little amalgam. The total sampling period is 24hrs with flow rate 0.25 (m^3/min). The total TGM concentrations were sum up by adding these from little amalgam after 24hrs sampling period.

Figure 2-2 displayed denuder sampling device. The heating system inside the insulated box consisted of a pair of general radiant heaters mounted inside the case and a pair of electronic temperature controlled (proportional, integral, and derivative PID) heating sleeves that were placed directly over the denuders to maintain 50°C. The denuder heating sleeves were constructed of polypropylene pipe insulation lined with silicone heating tape. Maintaining the denuders and inlets well above ambient temperatures during sampling was critical to prevent hy-drolysis of the KCl coating surface as well as ensure quantitative transport of RGM through the inlet. The volu-metrically flow-controlled pump was set to maintain 0.01 (m^3/min) to ensure an impact or aerodynamic aerosol cut point of 2.5 μm. [21]The average sampling time period was 1440±60 min for each sampling group. All the samples was collected and brought back to laboratory for analysis. Amalgam sampling quartz tube was connected to the Two-stage gold amalgamation by blowing the Ar gas into this amalgam sampling quartz tube. Cold-Vapor Atomic Fluorescence Spectrometry (CVAFS) was followed up for the RGM analysis in this study.

Figure 2-1 four-stage gold amalgamation sampling device

Figure 2-2 denuder sampling device

FIGURE 2-1 AMBIENT AIR TOTAL GASEOUS MERCURY (TGM) SAMPLING.
FIGURE 2-2AMBIENT AIR REACTION GASEOUS MERCURY (RGM) SAMPLING.

2.3 Cold-vapor atomic fluorescence spectrometry

Air was pulled through the vapor-phase sampling system using a mass-flow-controlled vacuum pump at a nominal flow rate of 0.3 L/min. Determination of vapor- and particle-phase mercury in ambient air was accomplished using dual-amalgamation Cold-Vapor Atomic Fluorescence Spectrometry (CVAFS) (Brook Rank, USA). The amount of vapor-phase mercury that was collected on a cold-coated bead trap was determined directly by CVAFS. The sample trap was heated to release the collected mercury. The desorbed mercury is carried in an inert gas stream (He or Ar) to a second cold-coated bead trap, which was the analytical trap. The mercury that is collected on the analytical trap is then thermally desorbed and carried into the CVAFS analyzer. The resulting voltage peak was integrated to determine the peak area for the sample.[22]

2.4 Quality control

TGM and RGM were blank test background contamination monitored by using operational blanks (unexposed glass fiber tube and glass fiber filter) which were processed simultaneously with field samples. Background contamination of mercury

was accounted for by subtracting field blank values from the concentrations. Field sample concentrations were obtained from subtracting blank samples for each sampling group. In this study, the background contamination is insignificant and can be ignored. The results average of the blank test was 1.96 pg for total gases mercury (TGM) and reaction gases mercury (RGM).

2.5 Mann-Whitney U test statistics

The Mann-Whitney U test is a non-parametric test that can be used in place of an unpaired t-test. It is used to test the null hypothesis that two samples come from the same population (i.e. have the same median) or, alternatively, whether observations in one sample tend to be larger than observations in the other. Although it is a non-parametric test it does assume that the two distributions are similar in shape. This study also applied Mann-Whitney U test to see the differences of average total gases mercury (TGM), reaction gases mercury (RGM) and gaseous element mercury(GEM) concentrations at traffic characteristic sample site.

III. RESULTS AND DISCUSSION

3.1 Meteorological conditions Hung-Kuang Traffic area sampling site during Dec. 2015 to Mar. 2016.

Table 1 present the meteorological conditions at the Hung-Kuang University Traffic area sampling sites from Dec 11, 2015 to Mar 16, 2016. The mean temperature, relative humidity, and wind speed (Dec 11, 2015 to Mar 16, 2016) at the Hung-Kuang University Traffic area sampling site were 17.06 ^{o}C, 76.81 %, and 4.7 m/s, respectively.

TABLE 1
ATMOSPHERIC METEOROLOGICAL CONDITIONS AT HUNG-KUANG UNIVERSITY TRAFFIC AREA DURING OFDEC. 2015 TO MAR. 2016.

Year	Sampling date	Temp. (℃)	R.H. (%)	W.S. (m/sec)	W.D.
2015	Dec 11'	20.12	76.20	4.8	NE
	Dec 14'	22.12	83.37	4.4	E
	Dec 16'	15.20	63.95	8.2	ENE
	Dec 18'	14.87	64.04	4.1	NE
	Dec 22'	22.29	88.75	2.4	SSW
	Dec 29'	17.41	70.54	3.6	SE
2016	Jan 12'	16.66	81.87	6.3	NE
	Jan 14'	14.58	78.12	5.7	E
	Jan 18'	14.54	72.12	5.7	E
	Feb 23'	14.33	82.95	7.3	ENE
	Feb 24'	13.47	78.86	6.8	ENE
	Mar 01'	14.41	62.70	3.8	SE
	Mar 03'	17.95	70.91	1.3	S
	Mar 07'	20.66	84.45	2.7	SSE
	Mar 16'	17.25	93.37	3.0	SE
	Mean	17.06	76.81	4.7	--

Temp:Temperature,RH: Relative humidity, WS: Wind speed, WD: Wind direction

3.2 Ambient air mercury species (TGM, RGM, GEM) concentrations during mid-winter～mid-spring in central Taiwan.

Table 2 display the TGM, RGM, GEM and RGM (%) concentrations during the year of Dec 11, 2015 to Mar 16, 2016. The result indicated that the highest average TGM concentration in Dec., 2015. In addition, the highest average RGM concentration in Mar.,2016. Moreover, the result indicated that the highest average GEM concentration in Dec., 2016. Meanwhile, the average RGM/TGM ratios were founded highest in March and the average ratios were 2.913. The possible reason is that the source surface emissions, such as ocean and related local industrial, activates were be in vogue during that period of time. And all the above industrial activate such as Taichung Thermal Power Plant (TTPP), Prowin Plastech co.,

LTD., King Kong Foundry co., LTD., were located in the western side of the sampling site. The wind was mainly direction blown the western side of the sampling site. Thus, high reaction gaseous mercury (RGM) concentration was measured during this period of time.

TABLE 2
AMBIENT AIR MERCURY SPECIES (TGM, RGM, GEM) CONCENTRATIONS DURING MID-WINTER～MID SPRING IN CENTRAL TAIWAN.

Year	Date	TGM	RGM	GEM	RGM (RGM/TGM)
		(pg/m^3)	(pg/m^3)	(pg/m^3)	(%)
2015	Dec 11'	6210	202.1	6010	3.25
	Dec 14'	8120	0.053	8120	0.001
	Dec 16'	8800	25.00	8770	0.28
	Dec 18'	6040	158.8	5880	2.63
	Dec 22'	9020	301.9	8720	3.35
	Dec 29'	10100	1.963	10080	0.02
	Average	8048	115.0	7930	1.589
2016	Jan 12'	10300	-	-	-
	Jan 14'	7740	3.265	7740	0.04
	Jan 18'	5870	238.8	5630	4.07
	Average	7970	121.0	6685	2.055
	Feb 23'	3900	1.163	3900	0.03
	Feb 24'	6840	1.123	6830	0.02
	Average	5370	1.143	5365	0.025
	Mar 01'	9220	32.26	9190	0.35
	Mar 03'	3710	47.32	3670	1.27
	Mar 07'	2490	67.58	2420	2.72
	Mar 16'	5100	372.8	4727	7.31
	Average	5130	129.99	5002	2.913

3.3 Comparisons with the other studies for TGM and RGM concentrations during years of 2010 ~ 2015

According reference of RGM concentrations may represent 1% of total gaseous Hg (TGM) at rural continental sites. (Ralph J. Valente et al, 2007) As for a traffic sampling site of this study, table 3 displayed average total gaseous mercury (TGM) and reaction gaseous mercury (RGM) concentrations with those of the other world regions during years of 2010-2015. The results indicated the average total gaseous mercury (TGM) and reaction gaseous mercury (RGM) concentrations were 6630 pg/m^3 and 91.8 pg/m^3 for this study, respectively and average total gaseous mercury (TGM) concentrations were 4280 pg/m^3 in the USA and China areas during years of 2010-2013. The average ratios were 0.65. Moreover, the average total gaseous mercury (TGM) concentrations were 3453 pg/m^3 in the Korea and Taiwan area during year of 2013. The mean ratios for ambient air mercury concentrations for Korea and Taiwan to this study were 0.52. In addition, the average reaction gaseous mercury (RGM) concentration was 2.35pg/m^3in the Canada and USA area during years of 2013-2014. The mean ratios for ambient air mercury concentrations for Canada and USA to this study were 0.025. Moreover, the average reaction gaseous mercury (RGM) concentrations were 23.91 pg/m^3 in the China, Korea and Spain area during years of 2011-2016. The mean ratios for ambient air mercury concentrations for China, Korea and Spain to this study were 0.26. To sum up, we conclude that the average highest TGM concentration was founded in Guiyang China. High economic and industrial which caused exhausted waste emissions were the possible reasons responsible for the above results. Moreover, Korea have the lower mercury species concentrations than that of China. On the contrary, the average TGM concentration measured in U.S.A were

lowest than those of the other Asian countries. Noteworthy, the average RGM concentration was founded in this study, central Taiwan. TTPP and THRIP which located in the western side of our sampling site. In addition, to TTPP and THRIP were mercury location such as Prowin Plastech co., LTD. and King Kong Foundry co., LTD. which concentration the potential sources for RGM. Thus, high RGM concentration was measured in this study. However, when compared with the other Asian countries, the results further indicated that Taiwan has the average highest RGM concentration followed by main land China during 2011~2015. And Korea has the lowest average RGM concentration. The average lowest RGM concentration were founded in U.S.A, Canada and Spain during years of 2013~2014. When compared with these of the other countries, the results showed that the U.S.A. has the average lowest mercury species (TGM, RGM, GEM) concentrations during the years of 2010～2013. The average TGM concentration ratio of China/ U.S.A. was about 2.5 in the year of 2010～2013.

TABLE 3
COMPARISONS WITH THE OTHER STUDIES FOR TGM AND RGM CONCENTRATIONS DURING YEARS OF 2010 ~ 2015

Country	Locations	TGM (pg/m^3)	Reference
USA	Detroit	2500	Liu et al., 2010
USA	Mississippi	2000	Jiang et al., 2013
China	Guiyang	9800	Fu et al., 2011
China	Waliguan	2000	Fu et al., 2012
China	Mt. Dinghu	5100	Chen et al., 2013
Koera	Guro-gu	3600	Kim et al., 2013
Taiwan	Taichung (HK Day)	3640	Fang et al., 2013
Taiwan	Taichung (HK Night)	3120	Fang et al., 2013
Taiwan	Taichung (Hung-Kuang)	6630	This study
Country	**Locations**	**RGM (pg/m^3)**	**Reference**
Canada	Halifax regional municipality	2.1	Cheng et al., 2014
Canada	Kejimkujik National Park	0.4	Cheng et al., 2014
USA	Huntington	1.3	Choi et al., 2013
USA	Rochester	5.6	Choi et al., 2013
China	Guiyang	35.7	Xuewu Fu et al., 2011
China	Xiamen	61.05	Lingling Xu et al., 2015
Korea	Seoul	11.3	Han et al., 2014
Korea	Chuncheon	2.7	Han et al., 2014
Spain	Puertollano	8.8	Alba Martínez-Coronado., 2016
Taiwan	Taichung (Hung-Kuang)	91.8	This study

3.4 Statistical (Mann-Whitney) analysis of TGM, RGM and GEM were the 24 hours period sampling site.

Table 4 displayed the statistical methods in the judging of the distribution differences for the ambient air TGM, RGM and GEM at Hung-Kuang Traffic Area sampling site. The results were summarized as followed:

Statistical analysis of correlation coefficients in TGM and RGM at the 24 hours period sampling site yielded a Mann-Whitney U statistic of 15.00, which is greater uα/0.05= 0.001, Mann-Whitney U of 0, suggesting that the sample population means are significant difference. Moreover, statistical analysis of correlation coefficients in RGM and GEM at the 24 hours period sampling site yielded a Mann-Whitney U statistic of 15.00, which is greater uα/0.05=0.001, Mann-Whitney U of 14.5, suggesting that the sample population means are significant difference.

TABLE 4
STATISTICAL (MANN-WHITNEY) ANALYSIS OF TGM, RGM AND GEM WERE THE 24 HOURS PERIOD SAMPLING SITE.

				Mann-Whitney	
Sampling sites	Mercury species	mean (pg/m^3)	SD	Mann-Whitney U statistic	P value
Hung-Kuang-Traffic -Area	TGM	6840	2414	0	0.001
	RGM	32.26	127.2		
	RGM	32.26	127.2	14.5	0.001
	GEM	6010	2325		

3.5 Relationship of concentration coefficients for ambient air TGM, RGM and GEM vs. temperature at Hung-Kuang traffic sampling site.

Figure 3 presents the correlation coefficients for ambient air TGM, RGM and GEM concentrations vs. temperature at Hung-Kuang traffic sampling site. TGM concentrations were ranged from 2490 to 10300pg/m^3, with a mean of 6629pg/m^3. RGM ranges from 0.053 to 372.8pg/m^3, with a mean of 91.78pg/m^3. GEM ranges from 2420 to 10080pg/m^3, with a mean of 6245pg/m^3. The results indicated that the correlation coefficients for TGM, RGM and GEM concentrations vs. temperature were 0.0009, 9E-06 and 0.0777, respectively. The above results indicated that TGM, RGM and GEM were low correlated to this of temperature at this traffic sampling site.

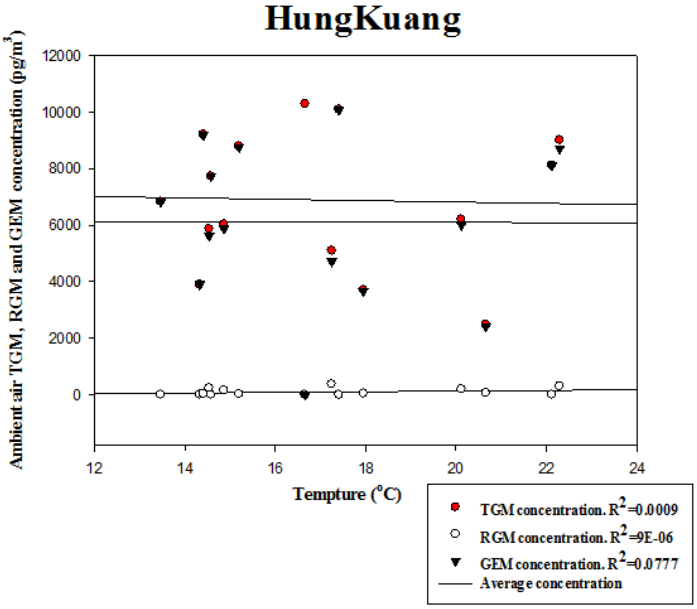

FIGURE 3 RELATIONSHIP OF CONCENTRATION COEFFICIENTS FOR AMBIENT AIR TGM, RGM AND GEM VS. TEMPERATURE AT HUNG-KUANG TRAFFIC SAMPLING SITE.

3.6 Relationship of concentration coefficients for ambient air TGM, RGM and GEM vs. wind speed at Hung-Kuang traffic sampling site.

Figure 4 presents the correlation coefficients for ambient air TGM, RGM and GEM concentrations vs. wind speed at Hung-Kuang traffic sampling site. TGM concentrations were ranged from 2490 to 10300pg/m^3, with a mean of 6629relationpg/m^3. RGM ranges from 0.053 to 372.8pg/m^3, with a mean of 91.78pg/m^3. GEM ranges from 2420 to 10080pg/m^3, with a mean of 6245pg/m^3.The results indicated that the correlation coefficients for TGM, RGM and GEM concentrations vs. wind speed

were 0.06, 0.0001 and 0.1539, respectively. The above results indicated that TGM, RGM and GEM were low correlated to this of wind speed at this traffic sampling site.

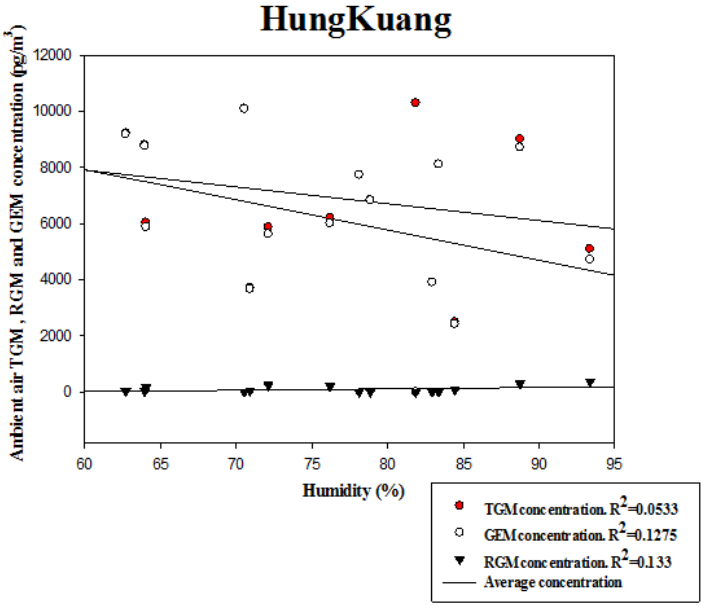

FIGURE 4 RELATIONSHIP OF CONCENTRATION COEFFICIENTS FOR AMBIENT AIR TGM, RGM AND GEM VS. WIND SPEED AT HUNG-KUANG TRAFFIC SAMPLING SITE.

3.7 Relationship of concentration coefficients for ambient air TGM, RGM and GEM vs. humidity at Hung-Kuang traffic sampling site.

Figure 5 presents the correlation coefficients for ambient air TGM, RGM and GEM concentrations vs. humidity at Hung-Kuang traffic sampling site. TGM concentrations were ranged from 2490 to 10300pg/m^3, with a mean of 6629pg/m^3. RGM ranges from 0.053 to 372.8pg/m^3, with a mean of 91.78pg/m^3. GEM ranges from 2420 to 10080pg/m^3, with a mean of 6245pg/m^3. The results indicated that the correlation coefficients for TGM, RGM and GEM concentrations vs. wind speed were 0.06, 0.0001 and 0.1539, respectively. Moreover, the above results further indicated that the higher the correlation humidity the lower the TGM, RGM and GEM concentrations were measured at this traffic sampling site. In contrast, the higher the relation humidity, the higher the RGM concentrations were measured at this traffic sampling site.

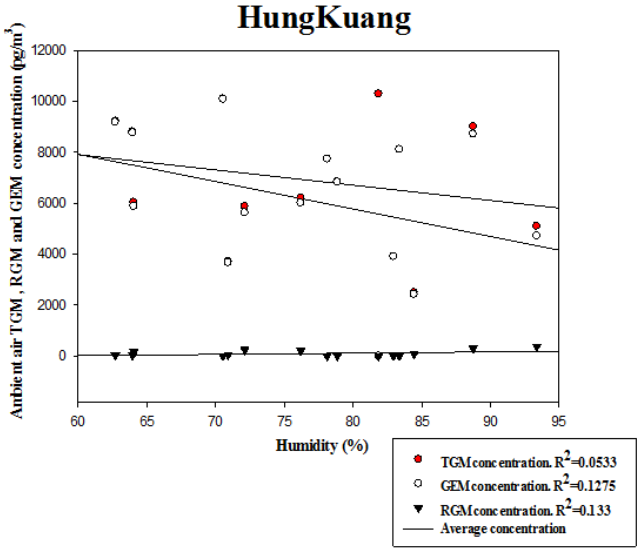

FIGURE 5 RELATIONSHIP OF CONCENTRATION COEFFICIENTS FOR AMBIENT AIR TGM, RGM AND GEM VS. HUMIDITY AT HUNG-KUANG TRAFFIC SAMPLING SITE.

IV. CONCLUSION

The major conclusions for this study are as follows:

- The average ratio for RGM concentrations out of the total gaseous mercury concentration was about 1.65% at the traffic sampling site.
- The average TGM concentration values obtained in this study were 6630 (pg/m^3) which is second to China average 9800 (pg/m^3) when compared with the other countries during year 2010～2013. In addition, Canada showed the average lowest RGM concentrations when Compared the other world areas during year of 2014～2015. The average RGM concentrations ratios for this study (average 91.8 pg/m3) to Canada were about 73.4 during years of 2014～2015.
- The results indicated that the correlation coefficients among total gaseous mercury (TGM), reaction gaseous mercury (RGM), gaseous element mercury GEM and meteorological conditions such as temperature and wind speed were low correlated related during mid-winter～mid-spring season at this traffic sampling site.
- Statistical method (Mann-Whitney U statistical) results revealed that there were significant differences in the mean concentrations values for the ambient air TGM, RGM and TGM were the Hung-Kuang sampling periods at this traffic sampling site.

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