Aerosol and Air Quality Research, 19: 375–382, 2019

ISSN: 1680-8584 print / 2071-1409 online
doi: 10.4209/aaqr.2018.11.0408

Air Pollution Profiles and Health Risk Assessment of Ambient Volatile Organic Compounds above a Municipal Wastewater Treatment Plant, Taiwan

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ABSTRACT

Municipal wastewater treatment processes have the function of removing harmful pollutants in the wastewater. However, there are probably several problems of air emissions related to these processes, especially for residents who live near a wastewater treatment plant. Volatile organic compounds exposure increases the risk of cancer. Thus, the health risk of residents to ambient volatile organic compounds exposure is essential to be conducted. One hundred and three volatile organic compounds (VOCs), total volatile organic compounds (TVOCs), and some prominent air pollutants (CO, CO_2, NH_3, H_2S, PM_1, $PM_{2.5}$, PM_7, PM_{10}, TSP) were investigated at the surface of an underground wastewater treatment plant in Taipei City during four different seasons. Twenty four VOCs were identified, some of which were categorized as carcinogenic to humans (Group 1) and possibly carcinogenic to humans (Group 2B) according to the International Agency for Research on Cancer. The mean values of CO, CO_2, PM_1, $PM_{2.5}$, PM_7, PM_{10} and TSP were found to be 0.64 ppm, 293.68 ppm, 1.37 μg m^{-3}, 3.20 μg m^{-3}, 10.74 μg m^{-3}, 13.48 μg m^{-3}, and 16.90 μg m^{-3}, respectively. NH_3 and H_2S were not detected in the present study. The health risk for residents was estimated following the method from United States Environmental Protection Agency (U.S. EPA). The cumulative of carcinogenic risk was 3.48×10^{-5} and categorized as a possible risk. In addition, the result was also possibly affected by traffic nearby. The magnitude for non-carcinogenic risk index was less than 1.

Keywords: Air pollutant; Wastewater treatment plant; Volatile organic compound; Cancer risk.

INTRODUCTION

Various types of contaminants are released into the atmosphere during the process of wastewater treatment and pollute the environment in numerous ways. There are odor problems as well as production of greenhouse gases during wastewater treatment (Hua *et al.*, 2018; Rai, 2018). Volatile organic compounds (VOCs) and other gaseous pollutants, such as methane, ammonia, hydrogen sulphide and particulate matter were detected in air surrounding the wastewater treatment plant (Hamoda, 2006; Widiana *et al.*, 2017).

Aeration and biological treatment have influences on the fates of aromatic volatile organic compounds (VOCs) in wastewater treatment processes (Chen *et al.*, 2013). VOCs in ambient air are an increasing concern because many of them have been identified to be human carcinogens (Chen *et al.*, 2016). Ammonia released into the atmosphere can constitute a source of olfactory nuisance (Chou and Wang, 2007). Hydrogen sulfide has peculiar smell and is also toxic to humans and environment (Liu and Wang, 2017). Particulate matter may affect the human respiratory, cardiovascular, and nervous system (Fang *et al.*, 2010; Haynes *et al.*, 2012; Wu *et al.*, 2013; Abe *et al.*, 2018; Zhang *et al.*, 2018).

Aeration process is one of the sources of particulate matter and NH_3 in wastewater treatment plant (Upadhyay

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et al., 2012). CO_2 is produced from biological wastewater treatment process and is defined as biogenic (Vijayan *et al.*, 2017). Moreover, other sources of ambient particulate matter was motor vehicle exhaust (Kumar and Yadav, 2016); therefore, if the location of the wastewater treatment plant is near the road, particulate matter will most likely be detected in its air surrounding. Solvent usages and paint applications are also sources of volatile organic compounds in municipal sewer and wastewater treatment plant (Huang *et al.*, 2012; Widiana *et al.*, 2017). While sources of ambient volatile organic compounds are mobile exhaust, stationary pollution from chemical or oil refinery plants, and natural emissions of animals and plants (Wang *et al*., 2016b).

Municipal wastewater treatment plant A is the largest secondary treatment plant in Taiwan with a capacity of 500,000 m^3 per day (TCG, 2017). It treats sewage from Taipei City household connections and interception stations. Most studies of air quality in wastewater treatment plants have focused mainly on the characteristics of microbial aerosols (Li *et al.*, 2013; Dehghani *et al.*, 2018). In the present study, the seasonal CO, CO_2, PM_1, $PM_{2.5}$, PM_7, PM_{10}, TSP, NH_3, H_2S, TVOC and VOCs concentrations in wastewater treatment plant A in Taipei City were measured outdoor. The TVOC seasonal concentration distributions were plotted using Surfer 10 program. This study was conducted on the surface of an underground municipal wastewater treatment plant A to investigate the air pollution profiles and the exposure level of residents nearby the wastewater treatment plant to volatile organic compounds. In addition, the health risks of ambient volatile organic compounds exposure for residents nearby the wastewater treatment plant were also estimated following the method from United States Environmental Protection Agency (U.S. EPA). Our findings provide a basis for improving the air quality in order to control the health risk of residents. The study of health risk assessment for residents nearby wastewater treatment plants is limited by the fact that few studies have done a similar investigation.

DATA AND METHODS

Sample Collection

All sampling collection was in the recreational sport park that was built on Municipal wastewater treatment plant A. The whole park was divided into four areas (A, B, C, and D area), and from each area, samples were taken at several points. The total sampling points for all areas were 30 (Fig. 1), while VOCs samples were taken at only one point for each of the four areas. The sampling was set at 1.2 m from the ground. Sampling periods were chosen in February 2016 (winter), May 2016 (spring), August 2016 (summer), and November 2016 (autumn). Samples were taken once in every season during the day of 08:00 am to 10:00 am.

Chemical Analysis

Sampling and its analysis were described in detail in a previous study (Widiana *et al.*, 2017). VOCs samples were collected using passive flow control canisters with volume 6 L and flow rate was fixed at 40 mL min^{-1}. All canisters were cleaned and vacuumed using humid N_2 pure gas to guarantee their vacuum quality before sampling. This study adopted the United States Environmental Protection Agency Method TO-15 and Photochemical Assessment Monitoring System for quality control during the sampling, preservation,

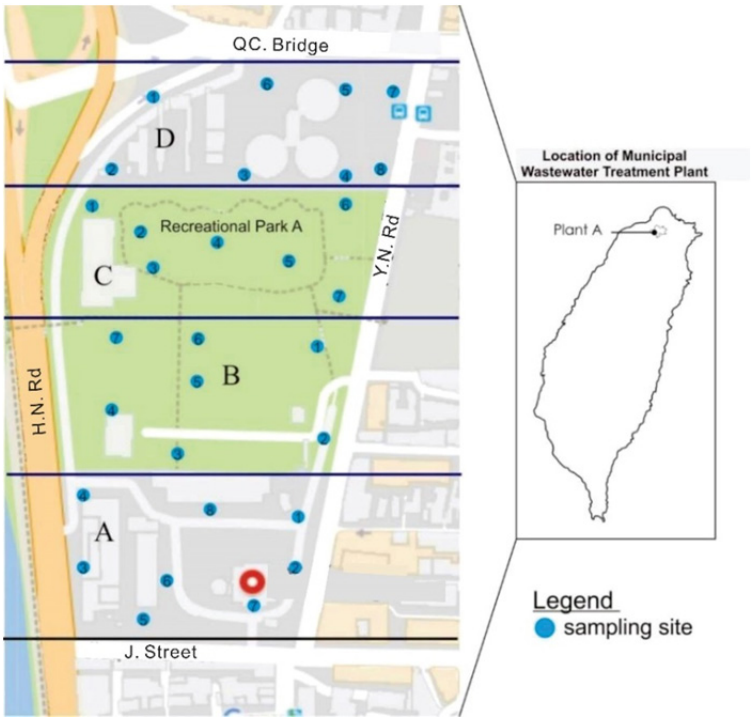

Fig. 1. Location of the sampling site.

transportation, and analysis. Air samples were then analyzed using a gas chromatograph (GC, Agilent 6890N) and a mass spectrometer (MS, Agilent 5973MSD). The GC oven temperature was set at 32°C, increased to 200°C, and kept constant for 3 min. Several standard gases were used to calibrate VOCs (Ou-Yang *et al.*, 2017). For each compound, calibration was done and a good linear fit was observed with $R^2 > 0.99$.

Sampling of CO and CO_2 was done using the instrument Q-TRAK™ Indoor Air Quality Monitor 7575 (TSI, Shoreview, United States). Calibration was done in the field. The appropriate detachable probes were attached to the instrument before field calibration, except for pressure and barometric pressure calibration. Particulate matter (PM) was measured using the instrument Met One Aerocet 531 particle profilers (Met One Instruments, Inc. Grants Pass, Oregon). The Aerocet 531 was calibrated using NIST (National Institute of Standard and Technology) traceable polystyrene. Real-time TVOC was measured using a portable PpbRAE 3000, photo-ionization detector (PID) having a 10.6 eV photoionization lamp detector (RAE System Inc., San Jose, CA). The TVOC monitor was calibrated using 100 ppm isobutylene and zero air following the manufacturer's recommendations (Singh *et al.*, 2016). The real ambient TVOC concentrations mapping were obtained using Surfer 10, which was developed by Golden Software Inc., USA. NH_3 and H_2S were measured using MultiRAE Lite PGM-620X (RAE Systems, San Jose, USA). Bump testing and gas detector calibration equipment were used regularly in accordance with OSHA guidelines. This is because bump testing equipment helps to ensure all the sensors are working properly to detect toxic gases.

Health Risk Assessment for the Residents

Health risk assessment focused on the chronic exposure to VOCs that are carcinogenic or non-carcinogenic, rather than acute exposure (He *et al.*, 2015). The lifetime cancer risk (LCR) is calculated using the equation (Singh *et al.*, 2016):

$$LCR = CDI \times CSF \quad (1)$$

Non-carcinogenic risk is characterized in terms of a hazard index (HI) which is defined as the ratio of chronic daily intake to the reference dose.

$$HI = \frac{CDI}{RfD} \quad (2)$$

While chronic daily intake (CDI) is calculated using the equation:

$$CDI = \frac{C \times CF \times IR \times EF \times ED}{BW \times AT} \quad (3)$$

The description of the variables used is summarized in Table 3. CSF and RfD were obtained by back-calculating the published unit risk and reference concentration values from U.S. EPA (U.S. EPA, 2017) based on the standard adult inhalation rate (20 m^3 day^{-1}) and lifetime (70 years) (Sofuoglu *et al.*, 2011), except for isoprene (Haney *et al.*, 2015). Table 4 lists the CSF and RfD.

RESULTS AND DISCUSSIONS

Level of Air Pollutants

Table 1 shows CO, CO_2, PM, TSP, H_2S, and NH_3 concentrations. The mean concentration of CO and CO_2 for four seasons were in the range 0.17–0.99 ppm and 259–310 ppm, respectively. The mean concentration of PM_1, $PM_{2.5}$, PM_7, PM_{10} and TSP for four seasons were in the range 0.53–1.67 μg m^{-3}, 2.10–4.97 μg m^{-3}, 7.43–17.9 μg m^{-3}, 9.33–22.5 μg m^{-3}, and 11.1–29.0 μg m^{-3}, respectively. In the present study, H_2S and NH_3 were not detected.

The results in the present study were close to previous study (Widiana *et al.*, 2017), except for $PM_{2.5}$, PM_7, PM_{10} and TSP which were lower. The previous study show the mean concentration of CO and CO_2 for four seasons in the ambient air were in the range 0–1.45 ppm and 286–322 ppm, respectively. The mean concentration of PM_1, $PM_{2.5}$, PM_7, PM_{10} and TSP for four seasons were in the range 0–2.09 μg m^{-3}, 2–6.64 μg m^{-3}, 8.55–22.6 μg m^{-3}, 10.2–27.1 μg m^{-3}, and 12–30.6 μg m^{-3}, respectively. H_2S and NH_3 were also not detected.

Hamoda (2006) investigated the presence of VOCs and other gaseous pollutants such as methane, ammonia, and hydrogen sulfide in air surrounding municipal wastewater

Table 1. CO, CO_2, PM, TSP, H_2S, and NH_3 concentrations in municipal wastewater treatment plant A.

Parameter	All			Winter			Spring			Summer			Autumn		
	n	Mean	SD	n	Mean	SD	n	Mean	SD	n	Mean	SD	n	Mean	SD
CO (ppm)	120	0.64	0.40	30	0.17	0.11	30	0.99	0.22	30	0.95	0.21	30	0.44	0.26
CO_2 (ppm)	120	293	33.5	30	259	15.0	30	310	33.2	30	304	34.3	30	298	21.4
PM_1 (μg m^{-3})	65	1.37	0.60	4	1.67	1.21	30	33.4	0.53	2	1	0	29	1	0
$PM_{2.5}$ (μg m^{-3})	118	3.20	1.60	28	3.39	0.88	30	3.30	0.70	30	2.10	0.84	30	4.97	1.81
PM_7 (μg m^{-3})	118	10.7	4.97	28	9.39	2.90	30	8.10	1.90	30	7.43	2.28	30	17.9	3.06
PM_{10} (μg m^{-3})	118	13.5	6.45	28	12.4	3.66	30	9.67	2.15	30	9.33	3.54	30	22.5	4.49
TSP (μg m^{-3})	118	16.9	10.1	28	15.4	5.09	30	11.1	2.42	30	12.0	5.97	30	29.0	11.6
NH_3 (ppm)	0	ND	-	0	ND	-	0	ND	-	0	-	ND	0	ND	-
H_2S (ppm)	0	ND	-	0	ND	-	0	ND	-	0	-	ND	0	ND	-

ND: not detected; n: number of samples; SD: standard deviation.

treatment plant in the State of Kuwait. In some cases the concentration exceeded the air quality standard. Lee *et al.* (2007) investigated the air quality of four wastewater treatment plants in Iowa, USA by monitoring the levels of hydrogen sulfide. The result show that the geometric means of hydrogen sulfide was less than 1 ppm.

Level of TVOCs

The locations of potential source of VOCs were identified using surfer program to plot seasonal concentration distributions. The concentration distribution of TVOCs for every seasons can be seen in Fig. 2. From Fig. 2, it can be seen that higher TVOC concentrations for each of the four seasons were distributed over the Sampling area A, which was surrounded by Y. N. Road, J. Street, and H. N. Road in the east, south, and west side, respectively. On the sampling area A, a car park can be found.

VOCs Characteristics

Benzene and toluene ratio (T/B) has been widely used as a simple method for evaluating the vehicle exhaust contribution to aromatics (Nelson and Quigley, 1984). T/B less than 2.0 indicated that aromatics were significantly influenced by vehicle emissions (Wang *et al.*, 2016a).

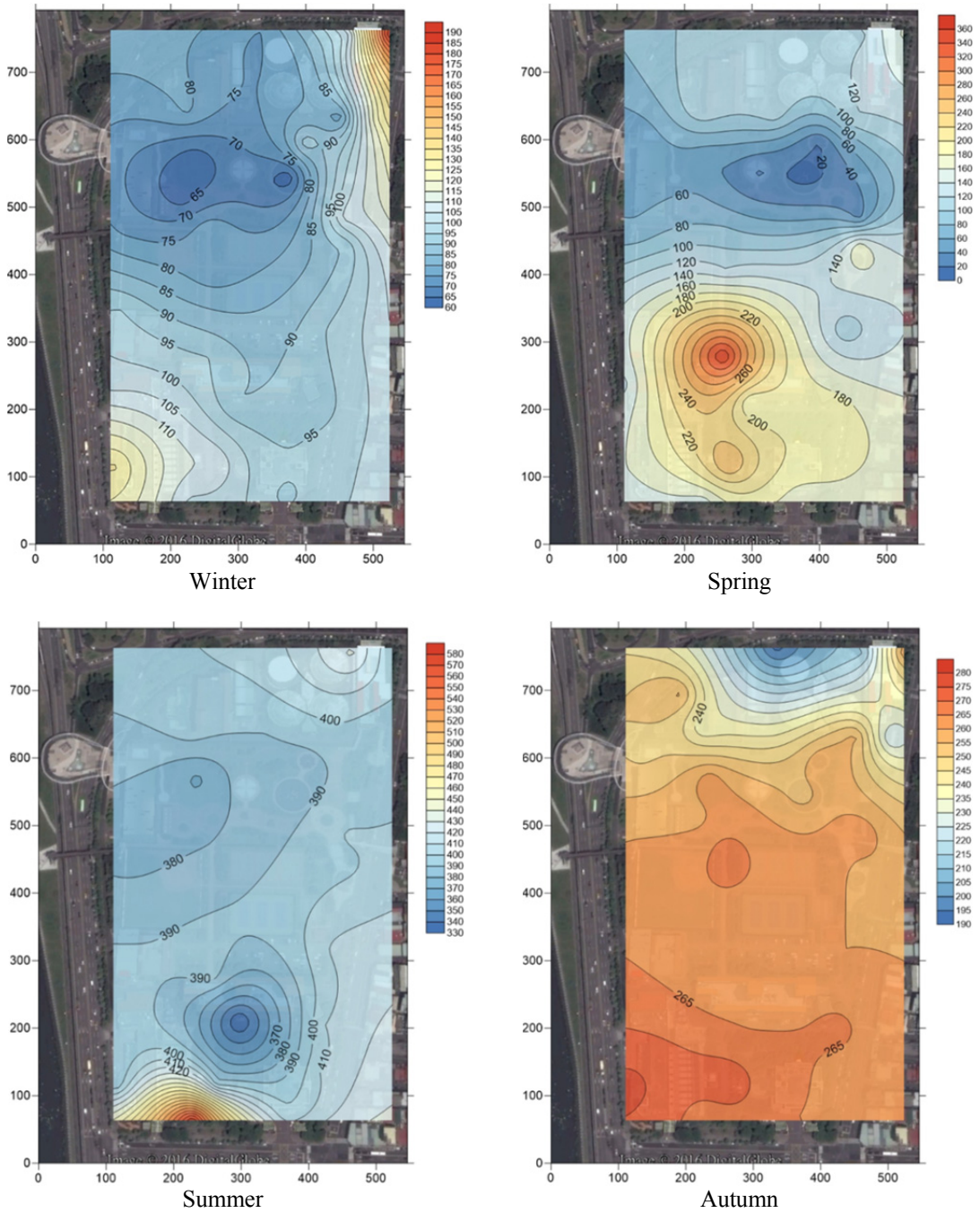

Fig. 2. Concentration distributions of TVOC for four seasons.

From Table 2, it can be seen that the average concentrations of toluene and benzene were 4.64 μg m^{-3} and 10.4 μg m^{-3}, respectively, therefore the T/B ratio obtained in the present study was 0.45, traffic influenced more. Ethanol yielded the highest mean concentration of VOCs followed by acetone and isopropyl alcohol.

Several factors that affect the seasonal variation of VOCs in the atmosphere were: first, the photochemical removal primarily by the hydroxyl (OH) radical in warmer seasons which result in higher chemical removal reaction rates than in cooler seasons because of more sunlight and high temperatures. Therefore the chemical removal of VOCs is faster in warmer seasons than in cooler seasons (Ho *et al.*, 2004). Second, the dilution as a result of atmospheric mixing. The dilution of airborne pollutants from ground water emissions in warmer seasons is stronger than in cooler system because the mixing layer is much higher in warmer seasons than in cooler seasons. Third, the principal source of VOCs, since the site is a recreational sports park, the principal source of VOCs will change with the tourism seasonal variations significantly.

Based on Table 2, the highest total of the VOCs appeared in spring because spring is a favorable season for tourism. Photochemical removal and dilution are weak because of the lower temperatures. Therefore, VOCs accumulates and the highest value appears in this season. Emission sources present in autumn are lower than those in summer and the effects of photochemical removal and dilution are still strong. Therefore, the lowest values of VOCs appeared in autumn (Zhang *et al.*, 2014). As shown in Fig. 3, the highest concentration and contribution of VOC groups was alkane then followed by alcohol and aromatics with concentration 50.7 μg m^{-3}, 47.6 μg m^{-3} and 34.9 μg m^{-3}, respectively.

Health Risk Assessment

In the present study, for all detected VOCs, only eight quantified VOCs were estimated according to the inhalation unit risk and reference dose values for carcinogenic and non-carcinogenic effects. Table 4 shows the lifetime cancer risk and hazard index. The lifetime cancer risk of the compounds determined by the US EPA was less than 10^{-6} and categorized as negligible or insignificant risk. The compounds with lifetime cancer risk less than 10^{-5} and higher than 10^{-6} were categorized as a possible risk, lifetime cancer risk less than 10^{-4} and higher 10^{-5} as a probable risk, and lifetime cancer risk higher than 10^{-4} as a definite risk (Sexton *et al.*, 2007).

According to the International Agency for Research on Cancer (IARC), three of the investigated VOCs are classified into two carcinogenic categories: group 1 (the agents is carcinogenic to human such as benzene) and group 2B (the agents is possibly carcinogenic to humans such as isoprene and methylene chloride). From Table 4 one compound (benzene) was regarded as possible risk and two other compounds (isoprene and methylene chloride)

Table 2. VOCs concentrations (μg m^{-3}) in municipal wastewater treatment plant A.

Compounds	All			Winter			Spring			Summer			Autumn		
	n	Mean	SD	n	Mean	SD	n	Mean	SD	n	Mean	SD	n	Mean	SD
Isobutane	7	6.95	3.40	3	5.07	3.50	1	4.14	0	1	10.3	0	2	9.50	1.43
n-Butane	5	13.6	4.22	1	9.92	0	1	17.4	0	1	18.7	0	2	10.9	1.60
Isopentane	16	12.9	7.16	4	11.3	5.96	4	10.3	9.13	4	14.2	7.51	4	15.9	7.41
n-Pentane	5	3.87	1.66	2	2.91	1.56	2	4.10	2.13	1	5.34	0	-	ND	-
2-Methylpentane	4	5.24	0.40	3	5.43	0.18	-	ND	-	-	ND	-	1	4.69	0
2-Methylhexane	1	5.37	0	1	5.37	0	-	ND	-	-	ND	-	-	ND	-
n-Hexane	1	2.78	0	1	2.78	0	-	ND	-	-	ND	-	-	ND	-
Benzene	4	10.4	13.50	4	10.4	13.50	-	ND	-	-	ND	-	-	ND	-
Toluene	12	4.64	4.12	4	4.29	3.26	4	7.36	5.65	1	4.41	0	3	1.56	0.44
m-Xylene	1	7.03	0	1	7.03	0	-	ND	-	-	ND	-	-	ND	-
1,2,4-Trimethylbenzene	1	12.8	0	1	12.83	0	-	ND	-	-	ND	-	-	ND	-
1-Hexene	1	5.4	0	-	ND	-	1	ND	-	1	5.40	0	-	ND	-
1-Butene	1	7.09	0	-	ND	-	-	ND	-	-	ND	-	1	7.09	0
Isoprene	3	4.46	1.40	-	ND	-	0	ND	-	3	4.46	1.40	-	ND	-
Methyl Ethyl Ketone	1	13.1	0	-	ND	-	-	13.1	0	-	ND	-	-	ND	-
Ethyl Acetate	1	13.9	0	-	ND	-	-	13.9	0	-	ND	-	-	ND	-
Isopropyl Alcohol	7	15.5	7.09	4	12.9	3.24	-	30.7	0	-	ND	-	2	13.2	1.72
Ethanol	16	32.0	19.33	4	22.6	9.64	4	27.0	22.6	4	34.5	13.20	4	44.0	27.2
Acetone	16	17.1	7.02	4	25.1	8.00	4	11.4	4.23	4	17.6	4.64	4	14.3	1.88
Methylene Chloride	5	5.34	2.60	4	4.96	2.84	1	ND	-	1	6.88	0	-	ND	-
Freon-12	10	5.98	1.57	-	ND	-	4	5.60	0.95	4	6.49	2.42	2	5.71	0.03
Freon-11	15	5.51	1.44	4	4.17	0.12	4	4.75	1.09	4	6.64	1.36	3	6.78	0.32
Freon-113	2	5.63	0.16	-	ND	-	1	5.52	0	1	5.75	0	-	ND	-
Freon-114	1	8.04	0	-	ND	3.50	-	8.04	-	1	ND	-	-	ND	1.43
Total		225			147			163			141			134	

ND: not detected; n: number of samples; SD: standard deviation.

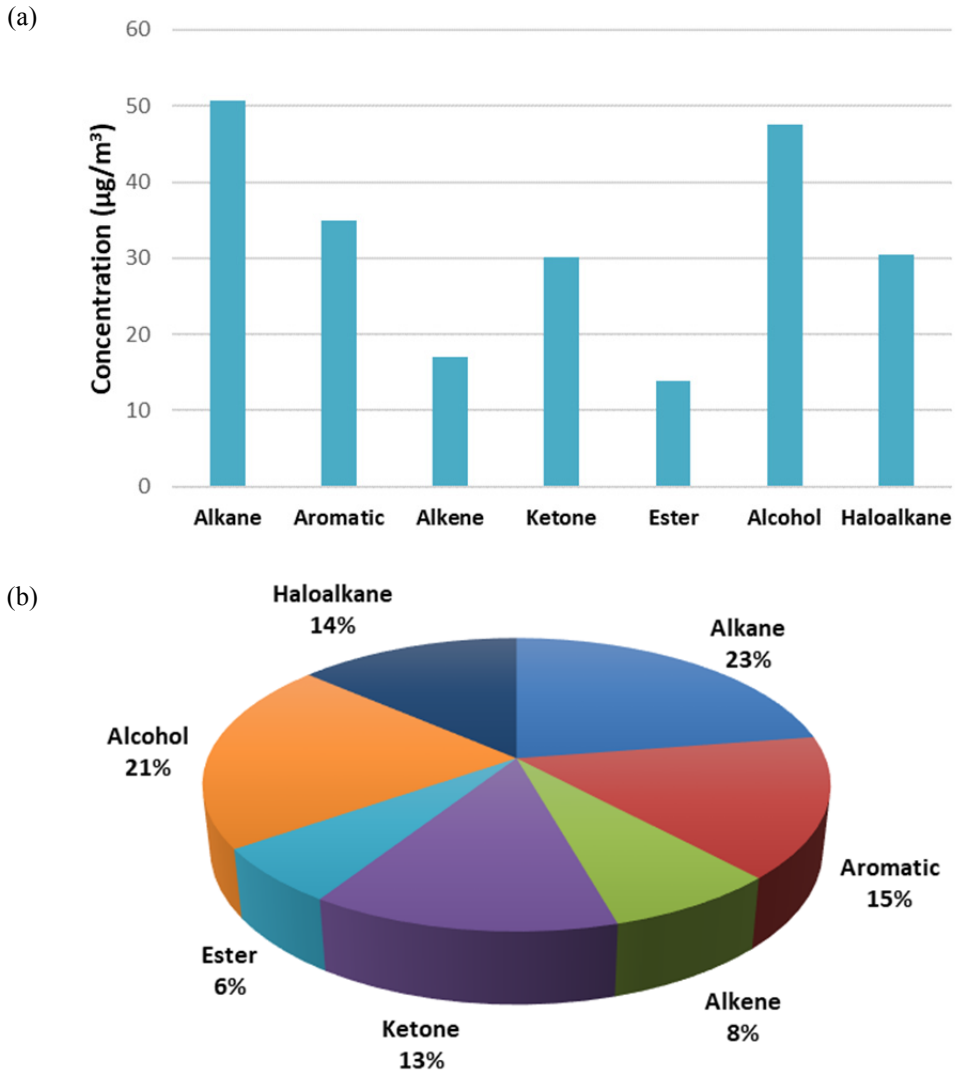

Fig. 3. (a) Concentration and (b) contribution of seven groups of VOCs.

were regarded as negligible or insignificant risk, and the cumulative of LCR was categorized as possible risk. Therefore, the provision of air pollution control is required to decrease the level of air pollutants. The identification of the air pollutant sources is the initial step in order to select the technology of air pollution control.

For the non-carcinogenic risk, HI greater than 1, the compound concentrations were considered to be above the level of concern (Ramírez *et al.*, 2012). Otherwise, it is assumed that the risk is at acceptable level (Biesiada, 2001). Nevertheless, for HI greater than 0.1 and lower than 1, the compounds were considered to be a potential risk to the residents health (McCarthy *et al.*, 2009). Table 4 shows the highest risk for non-carcinogenic originated from 1,2,4-trimethylbenzene. However, the cumulative of non-carcinogenic risk was less than 1 and considered unlikely to affect the residents.

One factor that affects the uncertainty of health risk assessment is VOCs concentration. However, the uncertainty was not analyzed in the present study due to the limitation in sampling size (Table 2). When the number of samples of VOCs is limited, then the distribution of VOCs concentration can not be accurately showed, which makes it difficult analyze the uncertainty. In future studies, the accuracy of the health risk assessment can be improved by considering a large size of samples.

CONCLUSIONS

Twenty four VOCs species, TVOCs, and some prominent air pollutants (CO, CO_2, NH_3, H_2S, PM_1, $PM_{2.5}$, PM_7, PM_{10}, TSP) were identified from municipal wastewater treatment plant A. Some VOCs were categorized as carcinogenic to humans (Group 1) and possibly carcinogenic to humans (Group 2B) according to the IARC. According to the results of this study, higher concentrations of TVOCs were in the sampling area A which was surrounded by three streets in the west, south, and east side. The carcinogenic risk for the residents was categorized as possible risk with value 3.48×10^{-5}. In addition, the hazard index value was 6.47×10^{-4} and considered unlikely to affect the residents.

Table 3. The exposure assessment factors.

Variable	Description	Value	Unit
C	Concentration		$\mu g\ m^{-3}$
CF	Conversion Factor	1000^{-1}	$mg\ \mu g^{-1}$
IR*	Inhalation Rate	20	$m^3\ day^{-1}$
EF	Exposure Frequency	365	$day\ year^{-1}$
ED	Exposure Duration	30	year
BW*	Body Weight	70	kg
AT	Average Lifetime		day
	- Carcinogenic	25550	
	- Non-carcinogenic	365 × ED	
CSF	Cancer Slope Factors		$(kg\text{-}day\ mg^{-1})$
RfD	Reference Dose		$(kg\text{-}day\ mg^{-1})^{-1}$

* U.S. EPA (1989).

Table 4. Carcinogenic (LCR) and non-carcinogenic risk (HI) of VOCs

Compounds	CSF	RfD	Chronic Daily Intake		Lifetime Cancer Risk	Hazard Index
			Carcinogenic	Non-carcinogenic		
n-Hexane[1]	-	200	-	7.96×10^{-4}	-	3.98×10^{-6}
Benzene[1]	0.027	8.57	1.27×10^{-3}	2.97×10^{-3}	3.48×10^{-5}	3.47×10^{-4}
Toluene[1]	-	1428	-	1.34×10^{-3}	-	9.28×10^{-7}
m-Xylene[1]	-	28.6	-	2.01×10^{-3}	-	7.03×10^{-5}
1,2,4-Trimethylbenzene[1]	-	17.1	-	3.68×10^{-3}	-	2.14×10^{-4}
Isoprene[2]	7.7×10^{-5}	-	5.46×10^{-4}	-	4.20×10^{-8}	-
Methyl Ethyl Ketone[1]	-	1429	-	3.74×10^{-3}	-	2.62×10^{-6}
Methylene Chloride[1]	3.5×10^{-5}	171	6.54×10^{-4}	1.53×10^{-3}	2.29×10^{-8}	8.90×10^{-6}
Total					3.48×10^{-5}	6.47×10^{-4}

[1] Reference concentration value and inhalation unit risk factor cited from US EPA's Integrated Risk Information System.
[2] Inhalation unit risk factor cited from Haney *et al*. (2015).

ACKNOLEDGEMENTS

The author thank to Chung Yuan Christian University for the support of this work

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Received for review, November 2, 2018
Revised, January 5, 2019
Accepted, January 15, 2019