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Excess Lifetime Cancer Risk Assessment of Volatile Organic Compounds Emitted from a Petrochemical Industrial Complex

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ABSTRACT

A health risk assessment for the Lin-Yuan Petrochemical Industrial Complex (LYPIC) in southern Taiwan was conducted in this study. The main aims of the study were to develop an emission inventory for the 21 processing plants in the LYPIC, conduct hazard identification based on the emission inventory, perform an exposure assessment by applying air dispersion modeling to obtain the annual average concentration of hazardous air pollutants near the LYPIC, and estimate the lifetime risk of cancer associated with volatile organic compound exposure for residents living in the vicinity of the LYPIC. Comparing with the reported monitoring data from several petrochemical and refinery plants in the world, the estimated exposure concentrations in this study were within reasonable range. The results of cancer risk assessment showed that the cumulative excess lifetime cancer risk (ELCR) at four representative residential sites surrounding the LYPIC were in the range of 9.3×10^{-5} to 1.7×10^{-4}. From a risk management perspective, in order to protect human health, greater emphasis on the reduction of emissions of benzene, 1,3-butadiene, acrylonitrile, and vinyl chloride is recommended. This study provides a feasible risk assessment procedure to identify the key carcinogenic chemicals emitted from a petrochemical industrial complex.

Keywords: Toxic air pollutants; Health effects/risks; Risk assessment; VOCs.

INTRODUCTION

The Lin-Yuan Petrochemical Industrial Complex (LYPIC) is in the Lin-Yuan district of Kaohsiung City in southern Taiwan. It is the second largest petrochemical industrial park in Taiwan, occupying an area of approximately 403 hectares and comprising 21 chemical and petrochemical companies. Many studies have indicated that the operation of petroleum refineries and petrochemical plants is associated with the generation of huge amounts of pollution, including SO_2, NO_x, respirable suspended particulate matter, polycyclic aromatic hydrocarbons (PAHs), and volatile organic compounds (VOCs) into the atmosphere (Pandya *et al.*, 2006; Rao *et al.*, 2008).

Petroleum refineries and petrochemical plants have been identified as the largest emitters of VOCs among the chemical industries (Pandya *et al.*, 2007; Baltrėnas *et al.*, 2011). In petrochemical plants, VOCs mainly originate from the production processes, storage tanks, transport pipelines, and waste areas (Kalabokas *et al.*, 2001; Cetin *et al.*, 2003; Ras *et al.*, 2009). Several environmental monitoring programs have been conducted worldwide to investigate the levels of VOCs in ambient air in the vicinity of petrochemical factories. The results of these studies showed that measured ambient VOC concentrations at vicinity of petrochemical complex and oil refinery were much higher than those measured at control sites (Na *et al.*, 2001; Cetin *et al.*, 2003). The results of a monitoring program conducted at the LYPIC indicated that maximum levels of a known human carcinogen, benzene, were as high as 618 ppbv during the day (Lin *et al.*, 2004). Another study in Alberta, Canada showed that 1,3-butadiene, another known human carcinogen, was measured at 27 ppbv downwind of an industrial area of more than 40 companies, including chemical, petrochemical, and oil and gas facilities (Simpson *et al.*, 2013). These results indicate that emissions of VOCs from petroleum refineries and petrochemical plants have a significant impact on surrounding areas.

VOCs in ambient air are an increasing concern because many of them have been identified to be human carcinogens

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(Simpson *et al.*, 2013). Occupational epidemiological studies of workers at petrochemical and refinery plants indicate that exposure to benzene has a dose-response relationship with the risk for acute non-lymphocytic/acute myelogenous leukemia (Bloemen *et al.*, 2004; Gun *et al.*, 2006; Kirkeleit *et al.*, 2008), exposure to vinyl chloride is associated with a substantial excess of angiosarcoma of the liver (Mundt *et al.*, 2000; Ward *et al.*, 2001), and exposure to butadiene is associated with an increased risk of non-Hodgkin lymphoma (Divine and Hartman, 2001). Two case-control studies from Taiwan indicated that living close to petrochemical industries may increase the risk of leukemia, especially in young people (Yu *et al.*, 2006; Weng *et al.*, 2008). However, a non-significant increase in the odds ratio for leukemia was obtained in the US (Tsai *et al.*, 2004), Italy (Belli *et al.*, 2004), and the UK (Sans *et al.*, 1995). As suggested by Barregard *et al.* (2009), this may be a result of limited information on exposure to carcinogenic VOCs.

Risk assessment is a tool used to organize, structure, and compile scientific information in order to identify existing hazardous situations, anticipate potential problems, establish priorities, and provide a basis for regulatory controls and/or corrective actions (World Health Organization, 2006). To the best of our knowledge, there is very limited research papers identifying all the species and amount of carcinogenic VOCs released or emitted from a petrochemical industrial complex. In addition, using four-step risk assessment methodology to estimate the lifetime risks of cancer associated with the carcinogenic VOCs exposures for residents living in the vicinity of a petrochemical industrial complex is rarely documented. However, carcinogenic VOCs released or emitted from a PC and the impact on the atmospheric environment within the vicinity are serious concerns. Therefore, the main aims of this study were to develop an emission inventory for the 21 processing plants in the LYPIC, conduct hazard identification based on the emission inventory, perform an exposure assessment by applying air dispersion modeling to obtain the annual average concentrations of VOC near the LYPIC, and estimate the lifetime risk of cancer associated with VOC exposure for residents living in the vicinity of the LYPIC.

MATERIALS AND METHODS

Defining the Scope of the Study Area

The LYPIC was established in the 1970s, and its processes can be divided into two basic categories: upstream and midstream. Basic petrochemical materials are produced from upstream sources, including ethylene, propylene, 1,3-butadiene, benzene, toluene, and p-xylene, while the midstream petrochemical intermediates, such as terephthalic acid, ethylene oxide, ethylene glycol, acrylonitrile, styrene, ethylbenzene, vinyl chloride, and acetaldehyde, are mostly derived from the basic petrochemical materials.

There are 24 villages in the Lin-Yuan district, which had a population of approximately 70,476 in 2014. As shown on the map in Fig. 1, the residential areas are located within 3.5 km of the LYPIC. We selected four representative villages, namely Site A, B, C, and D, as our target sites to estimate the lifetime risk of cancer associated with volatile organic compound exposure for residents living in the vicinity of the LYPIC.

Development of an Emission Inventory of Carcinogens from the LYPIC

There are 21 factories located in the LYPIC, which are expected to emit different kinds of VOCs. Four steps were performed in order to identify the species and emission rates of these chemicals. First, basic emission source data were collected for the 21 factories, including the processes used, the types of emission sources for these processes, and their permitted emission rates. The annual emission data was obtained from the permit system belonging to the local Environmental Protection Agency (EPA) of Kaohsiung City. Second, the actual annual emission rates for total volatile organic compounds (TVOCs) for each factory were determined after evaluating and comparing the annual emission rates from the Taiwan Emission Data Systems (TEDS) maintained by the Taiwan EPA. Third, the corresponding source profile for each of the VOC emission sources was selected from the US EPA SPECIATE 4.2 database (US EPA, 2009). By doing so, individual species of VOCs for each emission source could be identified. The emission rates could then be estimated and summed to obtain the total emission rate for each individual chemical from the LYPIC. Finally, all the chemicals were checked versus the International Agency for Research on Cancer (IARC) monograph on the evaluation of carcinogenic risks to humans. Those chemicals that were classified as carcinogenic to humans (Group 1), probably carcinogenic to humans (Group 2A), or possibly carcinogenic to humans (Group 2B) in the IARC monograph were targeted in this study. Table 1 provides a summary of reported annual emission rates for the 21 factories.

Atmospheric Dispersion Modeling

To establish the quantitative relationship linking the emission of VOCs from the LYPIC to their transport in the environment and exposure of target populations in the receiving areas, the study followed the framework recommended by the California Environmental Protection Agency (CALEPA, 2003). Most of the VOCs remain as gases when emitted into the air, and are not subject to appreciable deposition in soil, surface waters, or plants. Consequently, it is expected that the population would be exposed to VOCs primarily via ambient air. The relative contributions of routes of exposure other than the atmosphere (e.g., food intake, drinking water, and dermal contact) are known to be negligible. Therefore, when performing the risk assessment, estimation of the concentration of VOCs in the ambient air is the most important step (CALEPA, 2003).

In this study, ISCST3 (Industrial Source Complex-short term, Version 3) was used to obtain the annual average concentration of chemicals of concern at each site. The ISCST3 was used is due to the fact that the model is the regulatory air dispersion model suggested by Taiwan EPA. The simulation was performed using sequential hourly meteorological data obtained from the meteorological station

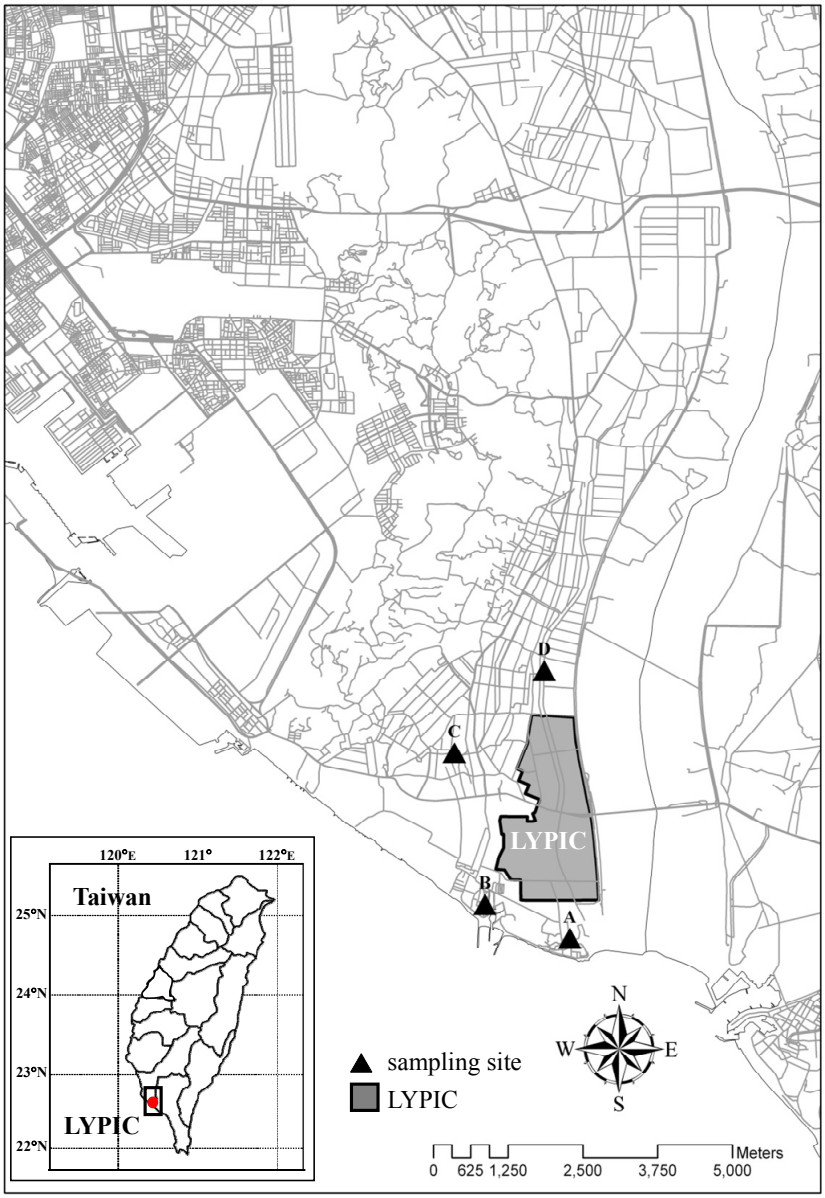

Fig. 1. The location of the LYPIC and four selected residential sites for estimation of cancer risk due to exposure to carcinogenic emissions.

near the study site for the 2005–2009. The dominant wind direction for the sampling site was from north to south (27%; Fig. 2). The annual average wind speed was 2.6 m s^{-1}.

Lifetime Cancer Risk Assessment

Excess lifetime cancer risk (ELCR) due to exposure through inhalation of carcinogenic VOCs emitted from the LYPIC was calculated by multiplying each estimated unit risk (UR) value by the annual average concentration of each chemical of concern as follows:

$$ELCR_i = C_i \times UR_i \quad (1)$$

where $ELCR_i$ is the ELCR value for chemical i, C_i is the annual average concentration of chemical i in ambient air (μg m^{-3}), and UR_i is unit risk value for i (μg m^{-3})$^{-1}$. The cancer UR describes the excess cancer risk associated with inhalation of a concentration of 1 μg m^{-3} of a given carcinogen.

The cumulative exposure to all the carcinogens emitted from the LYPIC was then summed to obtain the cumulative ELCR for residents living at a specific site by using the following formula:

$$Cumulative\ ELCR = \sum ELCR_i \quad (2)$$

RESULTS AND DISCUSSION

Summary of Emission Rates of TVOCs and Carcinogenic VOCs for the 21 Factories in the LYPIC

According to the statistical data from the Taiwan EPA, the reported average annual TVOCs emission rate for the

Table 1. The reported annual emission rates for TVOCs for the 21 factories in the LYPIC

Factory ID	Reported emission rate	
	Ton yr^{-1}	%
A	14.2	0.7
B	66.0	3.0
C	64.7	3.0
D	0.0	0.0
E	20.7	0.9
F	825.8	37.8
G	41.0	1.9
H	3.0	0.1
I	196.3	9.0
J	27.2	1.2
K	55.7	2.6
L	250.9	11.5
M	36.1	1.7
N	96.5	4.4
O	110.3	5.1
P	163.6	7.5
Q	83.0	3.8
R	52.3	2.4
S	5.9	0.3
T	53.8	2.5
U	16.8	0.8
Total	2183.8	100

LYPIC was 2183.8 tons in 2009 (as shown in Table 1). To identify the individual species of VOC emitted and determine the rates of emission for these chemicals each year from the factories in the LYPIC, data were collected from two datasets. The first dataset was the reported TVOC emission data submitted by the factories in the LYPIC (Table 1). The second dataset was extracted from SPECIATE 4.2 (US EPA, 2009), a database established by the US EPA, which provides VOC profiles for different manufacturing processes. SPECIATE 4.2 was used to estimate the number of individual VOC species and the amounts of each emitted when factories in the LYPIC engage in industrial manufacturing. Source profiles that corresponded to the industrial manufacturing processes of the LYPIC factories were selected according to the 58 emission classes in SPECIATE 4.2 and 5,187 source profiles. Altogether, 29 profiles were identified that were used in the LYPIC. The detailed of the manufacturing processes for each plant in LYPIC and their corresponding selected profiles were included in the Supplementary material. The percentage of each VOC emission constituent for the selected source profiles was multiplied by the total amount of each VOC emitted during industrial manufacturing processes to estimate the emission rate of each polluting chemical.

In total, 125 VOCs were identified as emissions from the LYPIC. The VOCs identified were then cross-referenced with the carcinogen classes defined in the IARC monograph to determine which chemicals should be included in the estimation of ELCR. Nineteen of the 125 VOCs were classified as Group 1, 2A, or 2B carcinogens. The 19 carcinogens and their estimated total emission rates from the LYPIC are shown in Table 2.

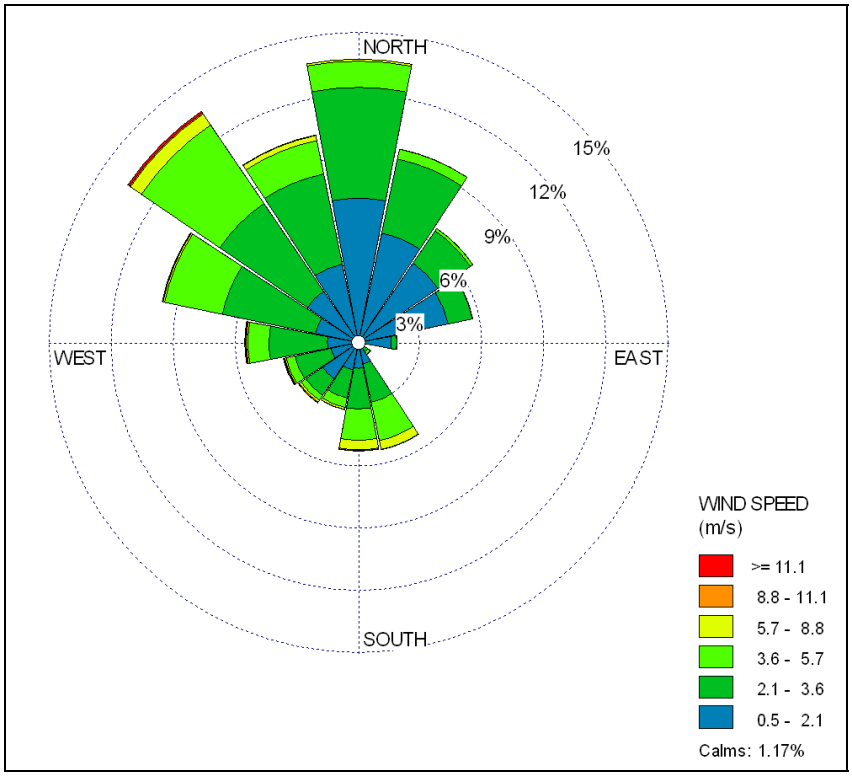

Fig. 2. Rose histogram depicting the distribution of wind direction and speed in the study area in 2005–2009. Wind direction is divided into 16 compass points. The percentages (circles) represent the occurrence of various wind speeds.

Table 2. Estimated emission rate of 19 carcinogens from the 21 factories in the LYPIC and unit risk (UR) values of these chemicals extracted from different toxicity databases

Chemical	IARC classification	ETAER[a] (ton yr^{-1})	Unit Risk (μg m^{-3})$^{-1}$			
			IRIS[b]	CALEPA[c]	Others	Value used in this study
1,3-Butadiene	1	22.3	3.0×10^{-5}	1.7×10^{-4}		3.0×10^{-5}
Benzene	1	117.7	7.8×10^{-6}	2.9×10^{-5}		7.8×10^{-6}
Ethylene oxide	1	0.05	3.7×10^{-6}	8.8×10^{-5}		3.7×10^{-6}
Formaldehyde	1	2.6	1.3×10^{-5}	6.0×10^{-6}		1.3×10^{-5}
Vinyl Chloride	1	44.6	8.8×10^{-6}	7.8×10^{-5}		8.8×10^{-6}
Trichloroethylene	1	0.3	4.1×10^{-6}	2.0×10^{-6}		4.1×10^{-6}
Tetrachloroethlyene	2A	1.2	2.6×10^{-7}	5.9×10^{-6}		2.6×10^{-7}
1,2-Dichloroethane	2B	2.6	2.6×10^{-5}			2.6×10^{-5}
Acetaldehyde	2B	2.3	2.2×10^{-6}	2.7×10^{-6}		2.2×10^{-6}
Acrylonitrile	2B	12.1	6.8×10^{-5}	2.9×10^{-4}		6.8×10^{-5}
Carbon tetrachloride	2B	1.3	6.0×10^{-6}	4.2×10^{-5}		6.0×10^{-6}
Chloroform	2B	2.7	2.3×10^{-5}	5.3×10^{-6}		2.3×10^{-5}
Ethyl acrylate	2B	2.4			1.4×10^{-5} [d]	1.4×10^{-5}
Ethylbenzene	2B	42.4		2.5×10^{-6}		2.5×10^{-6}
Isoprene	2B	0.5			3.8×10^{-6} [f]	3.8×10^{-6}
Methylene chloride	2B	2.4	1.0×10^{-8}	1.0×10^{-6}		1.0×10^{-8}
Propylene oxide	2B	0.2	3.7×10^{-6}			3.7×10^{-6}
Styrene[e]	2B	41.2			1.6×10^{-7} [e]	1.6×10^{-7}
Vinyl acetate	2B	2.0			3.7×10^{-7} [f]	3.7×10^{-7}

[a] ETAER: Estimated total annual emission rate from LYPIC;
[b] IRIS: US EPA, Integrated Risk Information System;
[c] CALEPA: California Environmental Protection Agency, Office of Environmental Health Hazard Assessment (OEHHA);
[d] HEAST: USEPA, Health Effects Assessment Summary Tables (HEAST);
[e] Guo *et al.* (2004);
[f] This study.

Table 2 also lists the UR values of the chemicals extracted from numerous toxicity databases, including the Integrated Risk Information System (IRIS) (US EPA, 2014), the Office of Environmental Health Hazard Assessment (OEHHA) (CALEPA, 2003), and the Health Effects Assessment Summary Table (HEAST) (US EPA, 1997). The UR values for 12 chemicals differed across different toxicity databases. By referencing the proposals by Tam and Neumann (2004) and Ramírez *et al.* (2012), toxicity data based on the database in which they were collected were prioritized, with IRIS being the most prominent, followed by CALEPA and all of the other databases. The last column of Table 2 shows the UR values used in the present study to assess ELCR.

The UR value could not be found in any of the toxicity databases for styrene, isoprene, or vinyl acetate. Guo *et al.* (2004) employed an oral carcinogenic slope factor (CSF) of 5.7×10^{-4} (mg kg^{-1} day^{-1})$^{-1}$ for styrene to undertake risk assessment in their study. Accordingly, a conversion equation proposed by USEPA (2002) and CALEPA (2003) was employed to convert CSF into UR:

$$UR = CSF \cdot \frac{1}{BW} \cdot IR \cdot CF \tag{3}$$

where, BW represents body weight (kg), IR represents inhalation rate (m^3 day^{-1}), and CF represents a unit conversion factor (mg μg^{-1}). Based on USEPA (2002), the suggested value for BW was 70 kg, for IR was 20 m^3 day^{-1}, and for CF was 0.001 mg μg^{-1}. Therefore, the UR for styrene was calculated at 1.6×10^{-7}(μg m^{-3})$^{-1}$.

Unfortunately, no CSFs were discovered for isoprene or vinyl acetate. To determine the UR value for isoprene, data were examined from the dose-response assessment presented in the 2-year carcinogenesis study of isoprene by the US Department of Health and Human Services, National Toxicology Program (National Toxicology Program, 1999). The data were then simulated using the Benchmark Dose Software (US EPA) to obtain the lower confidence limit of the benchmark dose (BMDL; 46.7 mg kg^{-1} day^{-1}). However, this benchmark dose was the amount administered to rats and required conversion into a human-equivalent dose (HED). Therefore, the following equation was employed (Shin *et al.*, 2010):

$$\text{HED} = \text{BMDL}/\text{CF} \tag{4}$$

where, CF represents the conversion factor (CF = 6.2 when the experimental subject is a rat). Using this equation, the estimated HED was 7.5 mg kg^{-1} day^{-1}. At a benchmark dose response (BMR) setting of 10%, CSF = BMR/HED = 1.3×10^{-2} (mg kg^{-1} day^{-1})$^{-1}$. The CSF was then converted to a UR for isoprene using Eq. (3), which was 3.8×10^{-6} (μg m^{-3})$^{-1}$.

Similarly, a dose-response assessment conducted on rats revealed the $BMDL_{10}$ for vinyl acetate to be 477 mg kg^{-1} day^{-1}

(Umeda *et al.*, 2004), which was converted to HED = 76.9 mg kg^{-1} day^{-1}. At a BMR of 10%, the CSF was 1.3×10^{-3} (mg kg^{-1} day^{-1})$^{-1}$. The CSF was then converted to a UR for vinyl acetate using Eq. (3), which was 3.7×10^{-7} (μg m^{-3})$^{-1}$.

The most abundant carcinogen emitted by the factories was benzene (Table 2), which is a key component in petrochemical processes. Benzene can be refined from crude oil or condensed from natural gas and is widely used to synthesize the intermediates of ethylbenzene, cumene, cyclohexane, nitrobenzene, and alkylbenzene. In addition, benzene is a crucial antiknock agent in gasoline (Fustinoni *et al.*, 2012). Analysis showed that all 21 factories located in the LYPIC emitted benzene into the surrounding environment, particularly those that distill petroleum, and manufacture ethylbenzene and alkylbenzene. Benzene is classified as a Group 1 carcinogen for humans by the IARC. Therefore, the production of benzene during industrial manufacturing and its subsequent emission into the surrounding environment necessitates immediate attention.

Results of Air Dispersion Modeling

Based on the estimated emission results shown in Section 3.1, an atmospheric dispersion simulation was conducted using annual meteorological data collected in the LYPIC from 2005–2009. The simulation was used to determine the average annual concentrations of the emissions in four populated sites (Sites A, B, C, and D) surrounding the LYPIC. The results of the simulation are illustrated in Fig. 3.

Fig. 3 shows that benzene has the highest concentration in this study. The concentration of benzene was 8.2, 3.6, 3.1, and 3.7 μg m^{-3} at Sites A, B, C, and D, respectively. At present, the European Union requires an average annual atmospheric benzene concentration of less than 5 μg m^{-3} (European Commission, 2000), and Japan sets an air quality standard for atmospheric benzene of 3 μg m^{-3} (Laowagul and Yoshizumi, 2009). In contrast to these regulations, the simulation results indicate that the benzene concentrations in the ambient air of the LYPIC were higher.

Table 3 summarizes the reported concentrations of benzene, 1,3-butadiene, and vinyl chloride collected near a PC or petroleum refinery in other countries. These three chemicals are the most commonly seen VOCs in the ambient air of petrochemical industrial plants. In research conducted by Reiss (2006) in Houston, the center of the petrochemical industry in the US, 14 monitoring stations were established throughout the Houston area and found that atmospheric benzene concentrations were 1.4–6.7 μg m^{-3} (mean = 3.0 μg m^{-3}) and atmospheric 1,3-butadiene concentrations were 0.1–7.1 μg m^{-3} (mean = 1.3 μg m^{-3}). These results are similar to the simulation results obtained in the present study. Furthermore, Fustinoni *et al.* (2012) sampled the atmosphere at a site 2 km from the petrochemical industrial zone in Sardinia, Italy. The results of chemical analyses showed that the median concentration for benzene in that area was 9 μg m^{-3}, and the concentration range from the 5th to the 95th percentile was 4–19 μg m^{-3}. A study of the atmosphere surrounding a refinery in Corinthians, Greece, showed that the mean concentration for benzene was 5.3 μg m^{-3} (Kalabokas *et al.*, 2001). A study conducted in the Ulsan Industrial Zone, Korea, reported a mean benzene concentration of 6.7 μg m^{-3} (Na *et al.*, 2001). In another study conducted in the Mumbai Industrial Zone, India, reported a mean benzene concentration

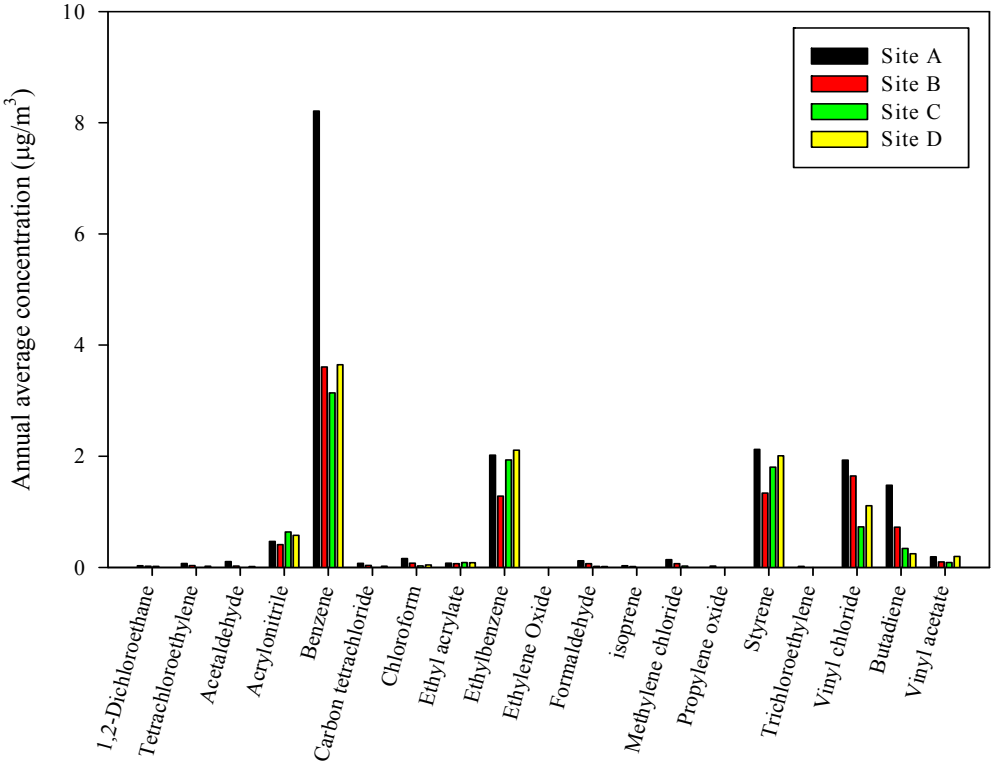

Fig. 3. Simulated annual average concentration of 19 chemicals at the four populated sites.

Table 3. Comparison of the simulated concentrations of benzene, 1,3-butadiene, and vinyl chloride in this study with reported values taken from within the vicinity of a petrochemical industrial complex or petroleum refinery.

City/Country	Site/chemical	Concentrations (μg m^{-3})	Source
Kaohsiung/Taiwan	LYPIC/Benzene LYPIC/1,3-butadiene LYPIC/Vinyl chloride	3.1–8.2 0.2–1.5 0.7–1.9	This study This study This study
Houston/USA	14 monitoring stations in the Houston area Benzene 1,3-butadiene	Mean: 3.0 Mean: 1.3	Reiss, 2006
Ulsan/Korea	Ulsan is one of the largest industrial areas in Korea with 700 petrochemical plants, oil refinery processes, and other chemical plants. Benzene Vinyl chloride	Mean: 6.7 Mean: 10.2	Na *et al.*, 2001
Bombay/India	Chemical and petrochemical industries in Bombay area Benzene	13.4–38.6	Rao *et al.*, 1997
Yokkaichi/Japan	One of the major industrial sources of 1,3-butadiene in Japan. 1,3-butadiene	1.7	Higashino *et al.*, 2007
Ichihara/Japan	Another major industrial sources of 1,3-butadiene in Japan. 1,3-butadiene	1.6	Higashino *et al.*, 2007

of 26.1 μg m^{-3} (Rao *et al.*, 1997). The results of these studies are consistent with the simulation results determined in the present study. It also indicated that the method we used to estimate the emission rate for individual chemical is reasonable to represent the situation for LYPIC.

1,3-butadiene is used to manufacture plastic and rubber, particularly for the production of polystyrene-butadiene-styrene (SBS) rubber, nitrile rubber, polybutadiene, acrylonitrile-butadiene-styrene (ABS), styrene, and acrylonitrile copolymer (SAN) resins (Dollard *et al.*, 2001). Higashino *et al.* (2007) used the METI-LIS model to estimate the 1,3-butadiene concentrations emitted into the surrounding atmosphere at three sites in Japan. They recorded a mean 1,3-butadiene concentration of 1.3 μg m^{-3} at a monitoring station in Yokkaichi yhogyokoko, 1.1 μg m^{-3} in Mihama shogakko, and 1.6 μg m^{-3} in Iwasakinishi. The atmospheric dispersion simulation results collected in the present study show that the 1,3-butadiene concentrations at the four sites were between 0.25 and 1.48 μg m^{-3}. Compared with the reported concentrations measured in Japan, these concentrations are considered to be within a reasonable range.

Vinyl chloride is a synthetic chemical used as an intermediate in the polymerization of polyvinyl chloride (PVC) in the petrochemical industry. High concentrations at petrochemical industrial complexes can be attributed to emissions from polymer industries. In the present study, the vinyl chloride concentrations for Sites A, B, C, and D were 1.93, 1.65, 0.73, and 1.11 μg m^{-3}, respectively. Na *et al.* (2001) reported vinyl chloride concentrations in the atmosphere surrounding the Ulsan Industrial Zone and in downtown Korea of 10.2 and 1.8 μg m^{-3}, respectively. Japan sets an annual average air quality standard for atmospheric vinyl chloride of less than 10 μg m^{-3} (Kawamoto *et al.*, 2011). The vinyl chloride concentrations collected at the four residential sites in this study were within this standard.

ELCR Assessment of Exposure to VOCs Emitted from the LYPIC

The simulated ELCR estimates for each carcinogen and cumulative ELCR estimates of all the carcinogens emitted from the LYPIC at the four representative neighboring residential sites are presented in Table 4. The cumulative ELCR estimates for Sites A, B, C, and D were 1.7×10^{-4}, 1.0×10^{-4}, 9.3×10^{-5}, and 9.3×10^{-5}, respectively. Currently, the target cumulative ELCR value set by numerous institutions, including the World Health Organization (2006), the Canadian Council of Ministers of the Environment (2006), and the New Zealand Ministry for the Environment (2010), is 1.0×10^{-5}. In Taiwan, Environmental Protection Agency set the target cumulative ELCR of 1.0×10^{-4}. The cumulative ELCR value of the carcinogens emitted from the LYPIC exceeded target values by 1.7-fold at site A. Therefore, we strongly recommend that the government in Taiwan employ relevant measures for risk management to ensure the health of residents living in the vicinity of the LYPIC and reduce the exposure associated with chemicals emitted from the LYPIC.

Based on the classification methods proposed in relevant literature (Morello-Frosch *et al.*, 2000), chemicals with an ELCR value less than 1.0×10^{-6} were categorized as posing an "uncertain risk", chemicals with ELCR values between 1.0×10^{-6} and 1.0×10^{-5} were categorized as posing a "possible risk", those between 1.0×10^{-5} and 1.0×10^{-4} were categorized as posing a "probable risk", and those greater than 1.0×10^{-4} were categorized as posing a "definite risk". For Site A, the site with the highest cumulative ELCR value, eight chemicals had ELCR values greater than $1.0 \times$

Table 4. Cumulative ELCR of this study and classification of the risk category for chemicals emitted from the LYPIC.

Chemical	ELCR$^{(\text{risk category})}$ (percent of contribution to cumulative ELCR)			
	Site A	Site B	Site C	Site D
1,3-Butadiene	$4.4 \times 10^{-5(B)}$ (26.0)	$2.2 \times 10^{-5(B)}$ (21.7)	$1.0 \times 10^{-5(B)}$ (11.1)	$7.4 \times 10^{-6(C)}$ (7.9)
Benzene	$6.4 \times 10^{-5(B)}$ (37.5)	$2.8 \times 10^{-5(B)}$ (28.1)	$2.4 \times 10^{-5(B)}$ (26.5)	$2.8 \times 10^{-5(B)}$ (30.5)
Ethylene Oxide	$1.3 \times 10^{-8(D)}$ (0.0)	$1.6 \times 10^{-9(D)}$ (0.0)	$2.2 \times 10^{-10(D)}$ (0.0)	$1.2 \times 10^{-9(D)}$ (0.0)
Formaldehyde	$1.5 \times 10^{-6(C)}$ (0.9)	$8.8 \times 10^{-7(D)}$ (0.9)	$2.4 \times 10^{-7(D)}$ (0.3)	$2.0 \times 10^{-7(D)}$ (0.2)
Vinyl chloride	$1.7 \times 10^{-5(B)}$ (9.9)	$1.4 \times 10^{-5(B)}$ (14.5)	$6.4 \times 10^{-6(C)}$ (6.9)	$9.8 \times 10^{-6(C)}$ (10.5)
Tetrachloroethylene	$1.8 \times 10^{-8(D)}$ (0.0)	$9.1 \times 10^{-9(D)}$ (0.0)	$3.4 \times 10^{-9(D)}$ (0.0)	$5.3 \times 10^{-9(D)}$ (0.0)
Trichloroethylene	$7.6 \times 10^{-8(D)}$ (0.0)	$3.8 \times 10^{-8(D)}$ (0.0)	$1.4 \times 10^{-8(D)}$ (0.0)	$2.2 \times 10^{-8(D)}$ (0.0)
1,2-Dichloroethane	$8.8 \times 10^{-7(D)}$ (0.5)	$5.9 \times 10^{-7(D)}$ (0.6)	$4.7 \times 10^{-7(D)}$ (0.5)	$1.6 \times 10^{-7(D)}$ (0.2)
Acetaldehyde	$2.3 \times 10^{-7(D)}$ (0.1)	$5.2 \times 10^{-8(D)}$ (0.1)	$1.8 \times 10^{-8(D)}$ (0.0)	$3.6 \times 10^{-8(d)}$ (0.0)
Acrylonitrile	$3.2 \times 10^{-5(B)}$ (18.7)	$2.8 \times 10^{-5(B)}$ (27.8)	$4.3 \times 10^{-5(B)}$ (47.0)	$3.9 \times 10^{-5(B)}$ (42.0)
Carbon tetrachloride	$4.5 \times 10^{-7(D)}$ (0.3)	$2.2 \times 10^{-7(D)}$ (0.2)	$8.2 \times 10^{-8(D)}$ (0.1)	$1.3 \times 10^{-7(D)}$ (0.1)
Chloroform	$3.7 \times 10^{-6(C)}$ (2.1)	$1.8 \times 10^{-6(C)}$ (1.8)	$6.7 \times 10^{-7(D)}$ (0.7)	$1.1 \times 10^{-6(C)}$ (1.1)
Ethyl acrylate	$1.1 \times 10^{-6(C)}$ (0.6)	$9.3 \times 10^{-7(D)}$ (0.9)	$1.2 \times 10^{-6(C)}$ (1.3)	$1.2 \times 10^{-6(C)}$ (1.3)
Ethylbenzene	$5.1 \times 10^{-6(C)}$ (3.0)	$3.2 \times 10^{-6(C)}$ (3.2)	$4.8 \times 10^{-6(C)}$ (5.2)	$5.3 \times 10^{-6(C)}$ (5.7)
Isoprene	$1.2 \times 10^{-7(D)}$ (0.1)	$5.9 \times 10^{-8(D)}$ (0.1)	$2.8 \times 10^{-8(D)}$ (0.0)	$2.0 \times 10^{-8(D)}$ (0.0)
Methylene chloride	$1.4 \times 10^{-9(D)}$ (0.0)	$7.0 \times 10^{-10(D)}$ (0.0)	$2.6 \times 10^{-10(D)}$ (0.0)	$4.1 \times 10^{-10(D)}$ (0.0)
Propylene oxide	$8.7 \times 10^{-8(D)}$ (0.1)	$2.0 \times 10^{-8(D)}$ (0.0)	$1.5 \times 10^{-9(D)}$ (0.0)	$4.8 \times 10^{-9(D)}$ (0.0)
Styrene	$3.4 \times 10^{-7(D)}$ (0.2)	$2.1 \times 10^{-7(D)}$ (0.2)	$2.9 \times 10^{-7(D)}$ (0.3)	$3.2 \times 10^{-7(D)}$ (0.3)
Vinyl acetate	$7.1 \times 10^{-8(D)}$ (0.0)	$3.7 \times 10^{-8(D)}$ (0.0)	$3.3 \times 10^{-8(D)}$ (0.0)	$7.4 \times 10^{-8(D)}$ (0.1)
Cumulative ELCR	$1.7 \times 10^{-4(A)}$ (100)	$1.0 \times 10^{-4(A)}$ (100)	$9.3 \times 10^{-5(B)}$ (100)	$9.3 \times 10^{-5(B)}$ (100)

Classification of risk category for chemical emitted from LYPIC:
(A): Definite risk;
(B): Probable risk;
(C): Possible risk;
(D): Uncertain risk.

10^{-6}. In sequential descending order, the chemicals were benzene (6.4×10^{-5}), 1,3-butadiene (4.4×10^{-5}), acrylonitrile (3.2×10^{-5}), vinyl chloride (1.7×10^{-5}), ethylbenzene (5.1×10^{-6}), chloroform (3.7×10^{-6}), formaldehyde (1.5×10^{-6}), and ethyl acrylate (1.1×10^{-6}). Among the eight chemicals, 1,3-butadiene, benzene, acrylonitrile, and vinyl chloride achieved an ELCR value greater than 1.0×10^{-5}, representing "probable risk" for the surrounding residents, whereas the remaining four chemicals posed a "possible risk." Moreover, the collective contribution of the eight carcinogens towards the overall cancer risk at Site A was approximately 98.7%. Therefore, controlling or reducing the emissions of these chemicals is essential for reducing the cancer risk for the residents in this area.

Comparison of Cancer Risk Assessment of Exposure to Carcinogens Emitted from Petrochemical Industrial Complexes

The present study estimated the health risks associated with the pollutants emitted from the LYPIC, a composite petrochemical industrial complex that encompasses upstream and midstream factories that engage in a variety of manufacturing processes. Therefore, estimation of emission of VOCs from the LYPIC is challenging. The approach we applied in this study provided a feasible methodology to identify the composition of VOCs as well as the emission rate of individual chemical emitted from the LYPIC.

In order to verify the reliability of ELCR estimated in the present study, we use the research results done by Ramírez *et al.* (2012) as reference for comparison. The results for both studies are summarized in Table 5.

In Ramírez *et al.* (2012), the authors selected the neighboring regions of two petrochemical industrial complexes located in Tarragona City, Spain, as the sampling sites, specifically, the North Industrial Complex and South Industrial Complex. They established three sampling sites at distances of between 500 and 1000 m from the complexes to sample the VOCs in the atmosphere. The cancer risk assessment showed that the estimated ELCR values for the three sites were between 3.2×10^{-4} and 3.4×10^{-4}. The area of the LYPIC was considerably smaller than the complexes of Tarragona City (403 ha vs. 470 ha + 717 ha). The overall emission of VOCs at the LYPIC (including methane) was estimated to be 2,183 tons yr^{-1}, with methane emissions comprising approximately 75 tons yr^{-1}. By contrast, the combined emission of non-methane VOCs sampled at the two Spanish complexes was 3,100 tons yr^{-1}. The meteorological conditions in the two studies were fairly similar. The present study recorded an average annual wind velocity of 2.6 m sec^{-1}, whereas Ramírez *et al.* (2012) recorded wind velocities of < 3 m sec^{-1}. Because of atmospheric dispersion, both studies set the distance between the sampling point and the emission source, which was within a comparable range of within 1,100 m. In this study, we used the ISCST3 to perform air dispersion modeling, where estimated emissions data and long-term meteorological data were collected and used to simulate the average annual concentrations in the nearby environment. By contrast, the

Table 5. Comparison of the ELCR estimated in this study with Ramírez *et al.* (2012).

	This study	Ramírez *et al.* (2012)
Site/Country	Lin-Yuan/Taiwan	Tarragona city/Spain
Area (ha)	403	Two Industrial Complex North: 470 South: 717
VOCs emission rate	2,183 ton yr^{-1} [a]	Non-methane VOCs 3,100 ton yr^{-1}
Analysis of VOCs concentrations	Source emission rate/ISCST3 air dispersion modeling	Samples were collected by means of active sampling with standard Orbo™-32charcoal tubes and then analyzed by GC/MS
Annual average wind speed	2.6 m sec^{-1}	< 3.0 m sec^{-1}
Distance of sampling site to industrial complex	Site A: 760 m Site B: 550 m Site C: 1110 m Site D: 300 m	Site 1: 500 m Site 2: < 500 m Site 3: < 1000 m
Simulated/measured annual VOCs concentrations at sampling site (μg m^{-3})	Site A Chloroform: 0.16 1,2-Dichloroethane: 0.03 Benzene: 8.21 Carbon tetrachloride: 0.07 Trichloroethylene: 0.02 Ethylbenzene: 2.02 Styrene: 2.12	Site 1 Chloroform: 0.65 1,2-Dichloroethane: 0.03 Benzene: 1.50 Carbon tetrachloride: 1.29 Trichloroethylene: 0.74 Ethylbenzene: 1.91 Styrene: 1.25
Excess lifetime cancer risk	Site A: 1.7×10^{-4} Site B: 1.0×10^{-4} Site C: 9.2×10^{-5} Site D: 9.3×10^{-5}	Site 1: 3.4×10^{-4} Site 2: 3.2×10^{-4} Site 3: 3.2×10^{-4}

[a] Emission rate of methane is estimated to be 75 ton yr^{-1}.

control study used the standard Orbo™-32 charcoal tube to collect 24-hour concentration values of 288 samples. These were converted into average annual concentration values. Analytical results show that the carcinogen concentrations obtained in the present study at Site A, which was 760 m from the LYPIC, were closely similar to those obtained by the comparator study at a monitoring site 500 m from the industrial complex in Spain. Specifically observing the concentrations for benzene, the present study obtained a concentration of 8.21 μg m^{-3}, whereas the comparator study obtained 1.50 μg m^{-3}. The reason for this 5.5-fold difference was primarily the result of various underlying conditions, such as factory processes, emission controls, and the type of petroleum industry. These differences may also be a result of uncertainty regarding elements generated during emission estimation. Moreover, the benzene concentrations obtained in the present study were also compared to those sampled near petroleum industrial complexes in other countries. A literature review showed that the ratio of benzene concentrations obtained in this study to Greece (Kalabokas *et al.*, 2001), Italy (Fustinoni *et al.*, 2012), and Korea (Na *et al.*, 2001) were 1.5, 0.9, and 1.2 μg m^{-3}, respectively. As these concentration ratios are all relatively close to 1.0, the simulation results of the present study can be considered to be within a reasonable range.

It is noteworthy that the comparator study employed an environment sampling method to collect VOC concentrations and subsequently used these concentrations to perform cancer risk assessments. However, carcinogen concentrations that are collected at these sampling sites may not originate from a single pollution source. For example, Ramírez *et al.* (2012) reported that the average daily automobile traffic flow at Sites 2 and 3 were 14,041 and 20,049 vehicles, respectively, suggesting that the concentrations collected at these sites also include those emitted from traffic source. By contrast, the present study performed health risk assessments by simulating atmospheric dispersion using the emissions data for the factories in the LYPIC. Essentially, the method employed in the present study assesses the cancer risks for the residents living in the vicinity of the LYPIC exposed to carcinogens emitted solely from the LYPIC. This assessment method was the primary distinction of the present study from the comparator study.

Uncertainty Analysis and Limitations of this Study

The LYPIC is a composite petrochemical industrial complex with more than 21 factories located in the complex. These plants used various manufacturing processes to produce upstream and midstream petrochemical products. Identification of the species of VOCs emitted from the LYPIC and estimation of the emission rates for these chemicals required using series of database. Inevitably, a level of uncertainty can be generated during these processes. In order to verify the estimated exposure level in the representative four sites in this study, we searched the reported air sampling data worldwide taken in the vicinity

of petrochemical industrial complexes (Rao *et al.*, 1997; Na *et al.*, 2001; Kalabokas *et al.*, 2001; Reiss, 2006; Higashino *et al.*, 2007; Fustinoni *et al.*, 2012) and compared with the results of simulation in this study. For example, the present study found that the primary contributive chemical to ELCR was benzene, following by 1,3-butadiene, and vinyl chloride. By comparing the simulated results with the sampling data of other studies (discussed in section 3.2 and section 3.4), we confirmed that the differences between the results of the present study and the sampling data in other reported studies were within reasonable ranges. A literature review showed that the ratio of benzene concentrations obtained in this study to Greece (Kalabokas *et al.*, 2001), Italy (Fustinoni *et al.*, 2012), and Korea (Na *et al.*, 2001) were 1.5, 0.9, and 1.2 μg m^{-3}, respectively.

During the cancer risk estimation process, we were not able to find the CSF values of two carcinogens (vinyl acetate and isoprene) from the toxicity database. To derive these UR values, we incorporated the dose-response data of test animals into the USEPA Benchmark dose software. The use of these estimates might produce a level of uncertainty in the final assessment results. However, it is noticed that the emission rates of vinyl acetate and isoprene only contributed to 0.7% and 0.2% of the overall carcinogen emission rates, respectively. Thus, the uncertainty generated from deriving the UR values for these two carcinogens should not create significant biases to the results of cancer risk assessment in this study.

In addition, the UR values obtained from IRIS and CALEPA showed considerable differences (Table 3). Using the more conservative UR values to calculate the ELCR for these 19 chemicals at Site A yielded a cumulative ELCR of 7.9×10^{-4}. While using the less-extreme UR values, the cumulative ELCR at Site A are estimated to be 1.7×10^{-4}. The difference is about 79%. The present study prioritized the UR values of IRIS over CALEPA, as suggested by numerous previous studies.

The present study employed the ISCST3 to simulate the atmospheric dispersion of VOCs. The ISCST3 is a non-reactive atmospheric dispersion model that overlooks the possibility that polluting chemicals may react during atmospheric dispersion. Numerous studies have found that chemicals such as 1,3-butadiene and vinyl chloride engage in photochemical reactions when exposed to sunlight, thereby reducing them in concentration (Dolye *et al.*, 2007; Liu *et al.*, 2013), and these reactions may cause simulations to produce overestimates. Studies have also confirmed that chemicals such as acrolein and formaldehyde are produced from the photochemical reaction of 1,3-butadiene (Baker *et al.*, 2005). Acrolein is classified as a Group 3 carcinogen (not classifiable as to its carcinogenicity to humans) by the IARC. Formaldehyde, which is also a chemical sampled at the LYPIC, may be underestimated due to photochemical reactions, resulting in the underestimation of cancer risk. By contrast, benzene is considered to be more stable in the atmosphere (Laowagul *et al.*, 2009). USEPA (2003) have conducted a study to compare the annual average concentrations obtained from ISCST3 modeling with observations from five field studies. The results showed that the modeling concentrations are 0.14-7.49 folds of the observations with an average of 2.4 (USEPA, 2003).

Finally, we used the approach of summing all the estimated individual cancer risk of exposure to each carcinogen to calculate the cumulative excess lifetime cancer risk for the selected residential sites in this study. Reports indicated that this approach does not reflect toxicological mechanisms of action (Anderson and Dennison, 2004). Possible mixture-related effects including antagonistic, synergistic, potentiating or additive effects may occur in complex mixtures (Ramírez *et al.*, 2012). Therefore, assuming components of a chemical mixture produce toxic effect through additivity may generate uncertainty. However, in the absence of data to the contrary, we followed the US EPA Guidelines for Carcinogen Risk Assessment (USEPA, 2005) to use the additivity model in this study.

CONCLUSIONS

The present study estimated the VOCs emitted from a petrochemical industrial complex, which included both upstream petroleum refining process and midstream petrochemical process. Based on the operating characteristics of the various factories in the LYPIC, 29 source profiles were screened from the SPECIATE 4.2 database. Among these profiles, a total of 125 species of VOCs were identified. Of these chemicals, 19 belonged to Group 1, 2A, or 2B carcinogens as classified by the IARC. Simulation results showed that the cumulative ELCR values at four representative residential sites surrounding the LYPIC, namely Sites A, B, C, and D, were 1.7×10^{-4}, 1.0×10^{-4}, 9.3×10^{-5}, and 9.3×10^{-5}, respectively. From a risk management perspective, the cumulative ELCR value of the pollutants emitted from the LYPIC exceeded the target value set by the Taiwan EPA. In this context, we strongly recommend that the government in Taiwan employ relevant measures for risk management to reduce health risks and thereby improve the health of residents living in the vicinity of the LYPIC. The simulation results obtained in the present study further suggest that greater emphasis should be placed on the emission of benzene, 1,3-butadiene, acrylonitrile, and vinyl chloride. These polluting chemicals are categorized as carcinogens classified as a "probable risk" in the present study, contributing 37.5% (benzene), 26.0% (1,3-butadiene), 18.7% (acrylonitrile), and 9.9% (vinyl chloride) to the ELCR value at Site A, which was the site with the greatest cancer risk. Overall, the present study not only evaluated the types, emission rates, and contributions to cancer risk of various carcinogens emitted from the factories of the LYPIC, but also compiled a list of polluting chemicals that require greater attention, which can serve as a valuable reference for decision makers. This study provides a rational and feasible risk assessment procedure for risk assessors and risk managers to conduct in complex emission situations.

ABBREVIATIONS

BMR, benchmark dose response; BW, body weight; CSF, carcinogenic slope factor; CF, conversion factor;

EPA, Environmental Protection Agency; EU, European Union; ELCR, excess lifetime cancer risk; HEAST, Health Effects Assessment Summary Table; HED, human-equivalent dose; ISCST3, Industrial Source Complex-short term, Version 3; IRIS, Integrated Risk Information System; IARC, International Agency for Research on Cancer; IR, inhalation rate; BMDL, benchmark dose limit; LYPIC, Lin-Yuan Petrochemical Industrial Complex; NATA, National-scale Air Toxics Assessment; OEHHA, Office of Environmental Health Hazard Assessment; PC, petrochemical complex; PAHs, polycyclic aromatic hydrocarbons; SBS, polystyrene-butadiene-styrene; PVC, polyvinyl chloride; SAN, styrene and acrylonitrile copolymer; TEDS, Taiwan Emission Data Systems; TVOC, total volatile organic compounds; UR, unit risk; VOC, volatile organic compound.

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DECLARATION OF COMPETING FINANCIAL INTERESTS

We have no actual or potential competing financial interests.

SUPPLEMENTARY MATERIALS

Supplementary data associated with this article can be found in the online version at http://www.aaqr.org.

REFERENCES

Andersen, M.E. and Dennison, J.E. (2004). Mechanistic approaches for mixture risk assessments—present capabilities with simple mixtures and future directions. *Environ. Toxicol. Pharmacol.* 16: 1–11.

Baker, J., Arey, J. and Atkinson, R. (2005). Formation and reaction of hydroxycarbonyls from the reaction of OH radicals with 1,3-butadiene and isoprene. *Environ. Sci. Technol.* 39: 4091–4099.

Baltrėnas, P., Baltrėnaitė, E., Šerevičienė, V. and Pereira, P. (2011). Atmospheric BTEX concentrations in the vicinity of the crude oil refinery of the Baltic region. *Environ. Monit. Assess.* 182: 115–127.

Barregard, L., Holmberg, E. and Sallsten, G. (2009). Leukaemia incidence in people living close to an oil refinery. *Environ. Res.* 109: 985–990.

Belli, S., Benedetti, M., Comba, P., Lagravinese, D., Martucci, V., Martuzzi, M., Morleo, D., Trinca, S. and Viviano, G. (2004). Case-control study on cancer risk associated to residence in the neighbourhood of a petrochemical plant. *Eur. J. Epidemiol.* 19: 49–54.

Bloemen, L.J., Youk, A., Bradley, T.D., Bodner, K. and Marsh, G. (2004). Lymphohaematopoietic cancer risk among chemical workers exposed to benzene. *Occup. Environ. Med.* 61: 270–274.

CALEPA (2003). Air Toxics Hot Spots Program Risk Assessment Guidelines. The Air Toxics Hot Spots Program Guidance Manual for Preparation of Health Risk Assessments. California Environmental Protection Agency. http://oehha.ca.gov/air/hot_spots/pdf/HRAguidefinal.pdf, Last access: 31 July 2014.

Canadian Council of Ministers of the Environment (2006). A protocol for the derivation of environmental and human health soil quality guidelines. Canadian Council of Ministers of the Environment.

Cetin, E., Odabasi, M. and Seyfioglu, R. (2003). Ambient volatile organic compound (VOC) concentrations around a petrochemical complex and a petroleum refinery. *Sci. Total Environ.* 312: 103–112.

Divine, B.J. and Hartman, C.M. (2001). A cohort mortality study among workers at a 1,3 butadiene facility. *Chem. Biol. Interact.* 135–136: 535–553.

Dollard, G.J., Dore, C.J. and Jenkin, M.E. (2001). Ambient concentrations of 1,3-butadiene in the UK. *Chem. Biol. Interact.* 135–136: 177–206.

Doyle, M., Sexton, K.G., Jeffries, H. and Jaspers, I. (2007). Atmospheric photochemical transformations enhance 1,3-butadiene-induced inflammatory responses in human epithelial cells: The role of ozone and other photochemical degradation products. *Chem.-Biol. Interact.* 166: 163–169.

European Commission (2000). Directive 2000/69/EC (2000) of the European Parliament and of the Council of 16 November relating to limit values for benzene and carbon monoxide in ambient air. Brussels, Belgium: European Commission.

Fustinoni, S., Campo, L., Satta, G., Campagna, M., Ibba, A., Tocco, M.G., Atzeri, S., Avataneo, G., Flore, C., Meloni, M., Bertazzi, P.A. and Cocco, P. (2012). Environmental and lifestyle factors affect benzene uptake biomonitoring of residents near a petrochemical plant. *Environ. Int.* 39: 2–7.

Gun, R.T., Pratt, N., Ryan, P. and Roder, D. (2006). Update of mortality and cancer incidence in the Australian petroleum industry cohort. *Occup. Environ. Med.* 63: 476–481.

Guo, H., Lee, S.C., Chan, L.Y. and Li, W.M. (2004). Risk assessment of exposure to volatile organic compounds in different indoor environments. *Environ. Res.* 94: 57–66.

Higashino, H., Mita, K., Yoshikado, H., Iwata, M. and Nakanishi, J. (2007). Exposure and risk assessment of 1,3-butadiene in Japan. *Chem. Biol. Interact.* 166: 52–62.

Kalabokas, P.D., Hatzaianestis, J., Bartzis, J.G. and Papagiannakopoulos, P. (2001). Atmospheric concentrations of saturated and aromatic hydrocarbons

around a Greek oil refinery. *Atmos. Environ.* 35: 2545–2555.

Kawamoto, T., Phan, T.T.P., Matsuda, T., Oyama, T., Tanaka, M., Yu, H.S. and Uchiyama, I. (2011). Historical review on development of environmental quality standards and guideline values for air pollutants in Japan. *Int. J. Hyg. Environ. Health* 214: 296–304.

Kirkeleit, J., Riise, T., Bråtveit, M. and Moen, B.E. (2008). Increased risk of acute myelogenous leukemia and multiple myeloma in a historical cohort of upstream petroleum workers exposed to crude oil. *Cancer Causes Control* 19: 13–23.

Laowagul, W. and Yoshizumi, K. (2009). Behavior of benzene and 1,3-butadiene concentrations in the urban atmosphere of Tokyo, Japan. *Atmos. Environ.* 43: 2052–2059.

Lin, T.Y., Sree, U., Tseng, S.H., Chiu, K.H., Wu, C.H. and Lo, J.G. (2004). Volatile organic compound concentrations in ambient air of Kaohsiung petroleum refinery in Taiwan. *Atmos. Environ.* 38: 4111–4122.

Liu, X., Yoon, S., Batchelor, B. andAbdel-Wahab, A. (2013). Photochemical degradation of vinyl chloride with an advanced reduction process (ARP) – Effects of reagents and pH. *Chem. Eng. J.* 215–216: 868–875.

Morello-Frosch, R.A., Woodruff, T.J., Axelrad, D.A. and Caldwell, J.C. (2000). Air toxics and health risks in California: The public health implications of outdoor concentrations. *Risk Anal.* 20: 273–291.

Mundt, K.A., Dell, L.D., Austin, R.P., Luippold, R.S., Noess, R. and Bigelow, C. (2000). Historical cohort study of 10109 men in the North American vinyl chloride industry, 1942–72: Update of cancer mortality to 31 December 1995. *Occup. Environ. Med.* 57: 774–781.

Na, K., Kim, Y.P., Moon, K.C., Moon, I. and Fung, K. (2001). Concentrations of volatile organic compounds in an industrial area of Korea. *Atmos. Environ.* 35: 2747–2756.

National Toxicology Program (1999). Toxicology and carcinogenesis studies of isoprene (CAS No. 78-79-5) in F344/N rats (inhalation studies). NTP TR 486, NIH Publication No. 99-3976. US Department of Health and Human Services, Public Health Services, National Insti tutes of Health. http://ntp.niehs.nih.gov/ntp/htdocs/lt_rpts/tr486.pdf, Last access: 19 September 2014.

New Zealand Ministry for the Environment (2010). *Draft toxicological intake values for priority contaminants in soil*. New Zealand Ministry for the Environment, Wellington.

Pandya, G.H., Gavane, A.G., Bhanarkar, A.D. and Kondawar, V.K. (2006). Concentrations of volatile organic compounds (VOCs) at an oil refinery. *J. Environ. Manage.* 63: 337–351.

Pandya, G.H., Kondawar, V.K. and Gavane, A.G. (2007). An integrated investigation of volatile organic compounds emission in the atmosphere from refinery and its off-site facilities. *Indian J. Chem. Technol.* 14: 283–291.

Ramírez, N., Cuadras, A., Rovira, E., Borrull, F. and Marcé, R.M. (2012). Chronic risk assessment of exposure to volatile organic compounds in the atmosphere near the largest Mediterranean industrial site. *Environ. Int.* 39: 200–209.

Rao, A.M.M., Pandit, G.G., Sain, P., Sharma, S., Krshnamoorthy, T.M. and Nambi, K.S.V. (1997). Non-methane hydrocarbons in industrial locations of Bombay. *Atmos. Environ.* 31: 1077–1085.

Rao, P.S., Ansari, M.F., Pipalatkar, P., Kumar, A., Nema, P. and Devotta, S. (2008). Measurement of particulate phase polycyclic aromatic hydrocarbon (PAHs) around a petroleum refinery. *Environ. Monit. Assess.* 137: 387–392.

Ras, M.R., Marce, R.M. and Borrull, F. (2009). Characterization of ozone precursor volatile organic compounds in urban atmospheres and around the petrochemical industry in the Tarragona region. *Sci. Total Environ.* 407: 4312–4319.

Reiss, R. (2006). Temporal trends and weekend-weekday differences for benzene and 1,3-butadiene in Houston, Texas. *Atmos. Environ.* 40: 4711–4724.

Sans, S., Elliott, P., Kleinschmidt, I., Shaddick, G., Pattenden, S., Walls, P., Grundy, C. and Dolk, H. (1995). Cancer incidence and mortality near the Baglan Bay petrochemical works, South Wales. *Occup. Environ. Med.* 52: 217–224.

Shin, J.W., Seol, I.C. and Son, C.G. (2010). Interpretation of animal dose and human equivalent dose for drug development. *J. Korean Orient. Med.* 31: 1–7.

Simpson, I.J., Marrero, J.E., Batterman, S., Meinardi, S., Barletta, B. and Blake, D.R. (2013). Air quality in the industrial Heartland of Alberta, Canada and potential impacts on human health. *Atmos. Environ.* 81: 702–709.

Tam, B.N. and Neumann, C.M. (2004). A human health assessment of hazardous air pollutants in Portland, OR. *J. Environ. Manage.* 73: 131–145.

Tsai, S.P., Cardarelli, K.M., Wendt, J.K. and Fraser, A.E. (2004). Mortality patterns among residents in Louisiana's industrial corridor, USA, 1970–99. *Occup. Environ. Med.* 61: 295–304.

Umeda, Y., Matsumoto, M., Yamazaki, K., Ohnishi, M., Arito, H., Nagano, K., Yamamoto, S. and Matsushima, T. (2004). Carcinogenicity and chronic toxicity in mice and rats administered vinyl acetate monomer in drinking water. *J. Occup. Health* 46: 87–99.

US EPA (1997). Health Effects Assessment Summary Tables (HEAST). EPA-540/R-97-036. Office of Solid Waste and Emergency Response, Washington, DC, USA, http://epa-heast.ornl.gov/index.html, Last access: 31 July 2014.

US EPA (2002). Industrial Waste Air Model Technical Background Document. EPA 530-R-02-010. Office of Solid Waste, Washington, DC, USA, http://www.epa.gov/solidwaste/nonhaz/industrial/tools/iwair/index.htm, Last access: 1 February 2015.

US EPA (2003). AERMOD: Latest features and evaluation results. EPA-454/R-03-003, June 2003, U.S. Environmental Protection Agency, Office of Air Quality Planning and Standards, Emissions Monitoring and Analysis Division, Research Triangle Park, NC 27711.

US EPA (2005). Guidelines for carcinogen risk assessment. Office of Research and Development, Washington, DC,

EPA/630/P-03/001B.

US EPA (2009). SPECIATE Database 4.2. http://cfpub.epa.gov/si/speciate, Last access: 1 February 2015.

US EPA (2014). Integrated Risk Information System (IRIS). http://www.epa.gov/IRIS/, Last access: 31 July 2014.

Ward, E., Boffetta, P., Andersen, A., Colin, D., Comba, P., Deddens, J.A., De Santis, M., Engholm, G., Hagmar, L., Langard, S., Lundberg, I., McElvenny, D., Pirastu, R., Sali, D. and Simonato, L. (2001). Update of the follow-up of mortality and cancer incidence among European workers employed in the vinyl chloride industry. *Epidemiology* 12: 710–718.

Weng, H.H., Tsai, S.S., Chiu, H.F., Wu, T.N. and Yang, C.Y. (2008). Association of childhood leukemia with residential exposure to petrochemical air pollution in Taiwan. *Inhalation Toxicol.* 20: 31–36.

World Health Organization (2006). *Evaluation of Certain Food Contaminations*. Sixty-fourth report of the joint FAO/WHO Expert Committee on Food Additives. WHO Technical Report Series 930. Geneva: World Health Organization.

Yu, C.L., Wang, S.F., Pan, P.C., Wu, M.T., Ho, C.K., Smith, T.J., Pothier, L., Christiani, D.C. and the Kaohsiung Leukemia Research Group (2006). Residential exposure to petrochemicals and the risk of leukemia: using geographic information system tools to estimate individual-level residential exposure. *Am. J. Epidemiol.* 164: 208–211.

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