

Article

# Indoor and Outdoor Exposure to Volatile Organic Compounds and Health Risk Assessment in Residents Living near an Optoelectronics Industrial Park

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**Abstract:** This study aimed to determine indoor and outdoor levels of volatile organic compounds (VOCs) and to assess potential risks among residents living in the vicinity of an optoelectronics industrial park in 2006–2007. We used steel canisters to collect 72 indoor samples and 80 outdoor samples over 24 h. Gas chromatography with a mass-selective detector was used for qualitative and quantitative analyses. The amounts of time residents spent doing activities in different microenvironments were determined by the self-administered questionnaire. The chronic hazard index (HIC) and cancer risk were applied to assess the non-carcinogenic and carcinogenic risks of VOCs among residents. Four VOCs of ethanol (indoor:  $77.8 \pm 92.8 \mu\text{g}/\text{m}^3$ ; outdoor:  $26.8 \pm 49.6 \mu\text{g}/\text{m}^3$ ), toluene ( $67.0 \pm 36.7 \mu\text{g}/\text{m}^3$ ;  $56.9 \pm 19.0 \mu\text{g}/\text{m}^3$ ), m/p-xylene ( $50.8 \pm 66.1 \mu\text{g}/\text{m}^3$ ;  $21.2 \pm 20.3 \mu\text{g}/\text{m}^3$ ), and acetone ( $37.7 \pm 27.5 \mu\text{g}/\text{m}^3$ ;  $25.8 \pm 9.8 \mu\text{g}/\text{m}^3$ ) were identified as dominant components in both the indoor and outdoor environments. Total VOCs and six VOCs of benzene, toluene, ethylbenzene, m/p-xylene, o-xylene, and ethanol in indoor sites were significantly higher than those in outdoor sites (all *p*-values < 0.05). All estimated HIC values were less than unity and the cancer risk of benzene exposure was  $1.8 \times 10^{-4}$  (range:  $9.3 \times 10^{-5}$  to  $3.4 \times 10^{-4}$ ) based on resident time-weighted patterns. Strategies to reduce benzene exposure should be implemented to protect public health.

**Keywords:** exposure assessment; health risk assessment; indoor air; optoelectronics industrial park; volatile organic compounds

## 1. Introduction

In order to revitalize economic growth, increase employment, and promote the upgrade of traditional industries, many emerging industrial parks have been established to construct a comprehensive chain of products for efficiency in developing countries. Among them, the Central Taiwan Science Park (CTSP), which has been in operation since July 2003, was built in the middle of Taiwan mainly to produce the thin-film transistor liquid-crystal display (TFT-LCD). According to the latest official statistics, this park, with a total area of 1486 hectares, includes 196 companies, 9 incubation centers, and 6 research units to attract the planned investment of \$697.3 billion in capital and the creation of 49,023 jobs by the end of March 2019 [1].

Exposure to volatile organic compounds (VOCs) has been reported to be associated with adverse health effects, including acute and chronic respiratory diseases, eye and throat irritation, neurological toxicity, kidney dysfunction, and lung cancer [2–9]. As the distance between the CTSP and the nearby

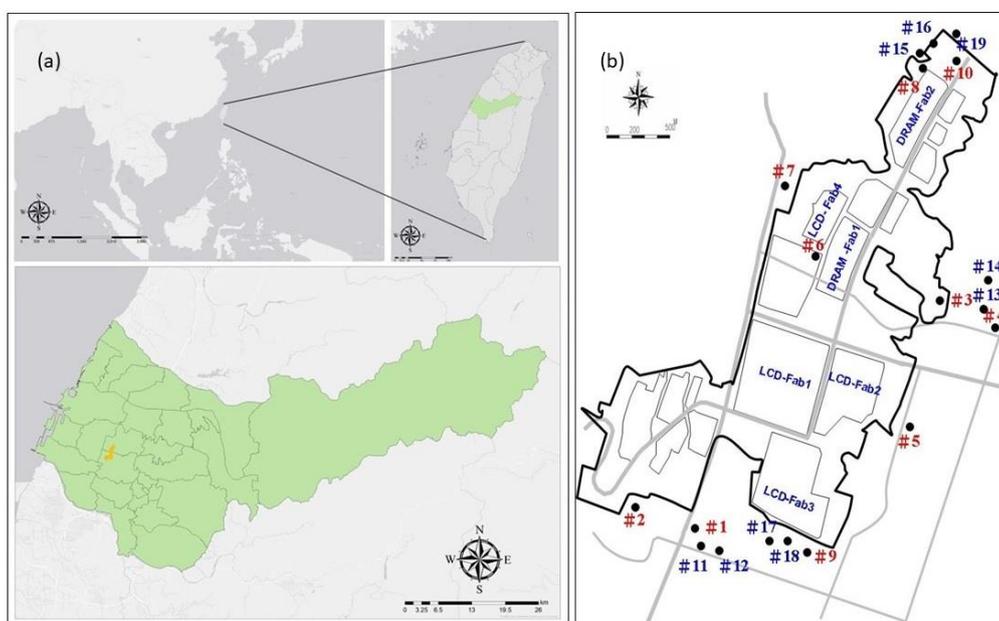
communities is short (less than 1 km), the emission of VOCs from industrial operations, which increases residents' exposure to air toxins, is of great concern. A previous study demonstrated the association between optoelectronics industrial activities and the increasing concentration of VOCs in the CTSP [10]. The concentrations of a total sum of 14 VOCs, toluene, acetone, and ethanol, were significantly associated with the increment of optoelectronic sales per million U.S. dollars after adjusting for traffic and meteorological conditions [10]. In the TFT-LCD companies, the measured VOCs included ethanol, acetone, isopropyl alcohol (IPA), propylene glycol monomethyl ether acetate (PGMEA), propylene glycol monomethyl ether, cyclohexanone, benzene, toluene, m/p-xylene, o-xylene, butyl acetate, and hexane [4,11,12]. Additionally, a very common chemical compound, tetramethylammonium hydroxide (TMAH), was reported to be used in the anisotropic wet etching of the TFT-LCD manufacturing processes [13].

To the best of our knowledge, no studies have been conducted to evaluate the potential health risks due to VOC exposure among inhabitants living in the vicinity of an optoelectronics industrial park. The purpose of this study was to determine indoor and outdoor levels of VOCs and to assess health risks among residents living near the CTSP over two years.

## 2. Materials and Methods

### 2.1. Study Site

The main base of the CTSP is located on the border between Situn District and Daya District of Taichung City in the middle of Taiwan, as shown in Figure 1a. There are about 325,300 people living in these two districts around the CTSP. The climate of the CTSP is a typical subtropical climate. During the study period, the average temperature ranged from 25.0 to 26.6 °C, the average humidity ranged from 66% to 75%, the average wind velocity ranged from 0.5 to 2.3 m/sec, and the prevailing wind was from the northwest direction (42% of the time in spring and summer and 77% in autumn and winter).



**Figure 1.** (a) Locations of the Central Taiwan Science Park (CTSP) in Taiwan; (b) outdoor (#1–#10, n = 10) and indoor (#11–#19, n = 9) sampling sites set up around the CTSP. DRAM—dynamic random access memory, LCD—liquid-crystal display.

### 2.2. Indoor and Outdoor Sampling

The sampling period was two years during 2006–2007. Each year covered four-day sampling to represent four seasons. Ten sampling sites were set up to collect ambient samples simultaneously from

07:30 to 07:30 over 24 h. As shown in Figure 1b, these outdoor sites included five sites located along four main roads to monitor both the traffic and industrial emissions of VOCs (i.e., #1, #4, #5, #7, and #10), four sites close to adjacent residential communities within 1.2 km of the park (such as #2, #3, #8, and #9), and one site with the estimated maximum concentration by the Industrial Source Complex (ISC) Short-Term 3 model (i.e., #6) to determine VOCs emitted mainly from industrial sources. Additionally, indoor samples were obtained from the living rooms of nine houses to estimate the inhalation exposure of residents to VOCs within 1.2 km of the park (i.e., #11–#19). The self-administrated questionnaire was also used to collect residents' daily time-activity patterns, lifestyle, and potential indoor sources of VOCs.

### 2.3. VOC Concentration Analysis

Integrated ambient air samples were collected in 6 L SUMMA stainless steel canisters with sillonite-coated valves (29-10622G model, Entech Instruments Inc., Simi Valley, CA, USA). Flow rates of 4.2 mL/min were used for 24-hour sampling. All canisters were cleaned using humid zero air with ten filling/evacuating cycles before sampling. One out of ten clean canisters was chosen to be analyzed to ensure that the concentration of all compounds inside a canister was below 10 parts per billion by volume (ppbv) and less than 0.2 ppbv for each of targeted VOC. Otherwise, the re-cleaning processes were performed for all canisters. Sampling and analysis of the samples followed the U.S. Environmental Protection Agency Compendium Method TO-15 [14]. An injection volume of 100 mL for VOCs sampled in the canister was concentrated in an ENTECH 7100A cryogenic concentrator and was injected into a HP6890 gas chromatograph coupled to a HP5973 mass-selective detector (GC/MS). The ENTECH 7100A concentrator first transferred the air sample through a liquid nitrogen cooled cryogenic trap of glass beads at  $-150\text{ }^{\circ}\text{C}$  for the removal of water. The trapped VOCs were then transferred by pure helium to a secondary Tenax TA trap at  $-30\text{ }^{\circ}\text{C}$  for the removal of  $\text{CO}_2$ . The secondary trap was heated ( $180\text{ }^{\circ}\text{C}$  for 3.5 min) to transfer the target VOCs to a third cryogenic trap at  $-160\text{ }^{\circ}\text{C}$ . Finally, the trap was rapidly heated to  $70\text{ }^{\circ}\text{C}$ , and then the VOCs were transferred to the GC/MS system. A capillary column (J&W DB-502.2, 60 m  $\times$  0.25 mm  $\times$  1.4  $\mu\text{m}$ ) was used in this system. The analysis started at  $35\text{ }^{\circ}\text{C}$  and immediately increased at a rate of  $5\text{ }^{\circ}\text{C}/\text{min}$  to  $90\text{ }^{\circ}\text{C}$ . The temperature was held at  $90\text{ }^{\circ}\text{C}$  for 2 minutes and then raised to  $170\text{ }^{\circ}\text{C}$  at a rate of  $7\text{ }^{\circ}\text{C}/\text{min}$  and kept at  $170\text{ }^{\circ}\text{C}$  for 3.3 minutes. Finally, the temperature was increased from  $170\text{ }^{\circ}\text{C}$  to  $210\text{ }^{\circ}\text{C}$  at a rate of  $10\text{ }^{\circ}\text{C}/\text{min}$  and held at  $210\text{ }^{\circ}\text{C}$  for 5 minutes. The carrier gas was helium at a flow rate of 1 mL/min. The split flow was set to give a split of 1:10 to avoid the peak broadening and tailing phenomenon. The MS was used to identify and quantify the eluted species based on each compound's retention time and fragmentation pattern. The operational conditions of MS were set to range from 33 to 300 amu with the electron multiplier at 1.1 kV. The scan interval was 0.5 seconds and the scan speed was 5 scans per second.

### 2.4. Quality Assurance/Quality Control

The laboratory and field blanks, parallel samples, and duplicate measurements of samples were subjected to quality assurance and quality control procedures. In the laboratory, a blank sample was randomly selected from ten canisters that had completed the cleaning procedure for analysis to ensure that the concentration of all compounds inside was below 10 ppbv and less than 0.2 ppbv of any targeted VOC. A field blank was analyzed on each sampling day. Parallel samples and duplicate measurements of samples were analyzed to test the precision of the sampling and analytical techniques, respectively. The mean relative standard deviations (R.S.D.) for all the compounds measured at the level of 10 ppbv were less than 10%. A new calibration curve was determined each time. The detection limit of each compound was calculated from the data of five replicate measurements of low concentration samples and observed from their standard deviation. The analyzed VOCs revealed the detection limits ranged from  $0.4\text{ }\mu\text{g}/\text{m}^3$  (ethanol) to  $0.9\text{ }\mu\text{g}/\text{m}^3$  (propylene glycol monomethyl ether acetate, PGMEA). Four internal standards (bromochloromethane, 1,4-difluorobenzene, chlorobenzene-d<sub>5</sub>, and 1-bromo-4-fluorobenzene) blended in for each injection aliquot were used in the calibration system to

confirm the stability of MS and to ensure data quality. The recovery difference between the standard gas and each target VOC was required to be less than 30%. The recovery rates of all VOCs analyzed ranged from 96.2% (acetone) to 101.1% (m/p-xylene).

### 2.5. Risk Estimates

Benchmark estimates of carcinogenic and noncarcinogenic risks for each VOC were taken directly from the California Environmental Protection Agency (Cal-EPA) [15,16]. The chronic reference exposure levels (REL) for airborne toxicants proposed by the Cal-EPA were used for comparison purposes [15]. The hazard index of chronic effects for the  $i$  VOC ( $Hlc_i$ ) was calculated as follows. We first calculated the indoor and outdoor time-weighted average concentration of the  $i$  VOC ( $\bar{C}_i$ ) over the study period using the following equation (1):

$$\bar{C}_i = \frac{Indoor_c \times Time_{indoor} + Outdoor_c \times Time_{outdoor}}{Time_{total}}, \quad (1)$$

where  $Indoor_c$  represents the two-year average concentration of nine indoor sites,  $Time_{indoor}$  is the average daily period spent in the indoor environment from the two-year questionnaire survey,  $Outdoor_c$  represents the two-year average concentration of 10 outdoor sites,  $Time_{outdoor}$  is the average daily period spent in the outdoor environment, and  $Time_{total}$  is 24 h. Second, the time-weighted average concentration of specific VOCs was divided by its respective chronic reference exposure level ( $REL_{c_i}$ ). Finally, we summed the  $\bar{C}_i/REL_{c_i}$  measurements of specific VOCs with the same adverse effects of target organs or systems to obtain  $Hlc_j$  for the  $j$  health effects, as shown in Equation (2). Only VOCs with RELc and measured levels above the limit of detection each season were used to calculate specific  $Hlc_j$  values. Residents living in the vicinity of this park had potential chronic health risks if the calculated  $Hlc_j$  of the specific adverse effect was greater than 1.

$$Hlc_j = \frac{\bar{C}_1}{REL_{c_1}} + \frac{\bar{C}_2}{REL_{c_2}} + \dots + \frac{\bar{C}_i}{REL_{c_i}}, \quad (2)$$

where  $i$  represents one of six VOCs with measurable REL, and  $j$  represents different health effects.

We assumed that each resident had a respiration rate of 20 m<sup>3</sup> per day and exposure to VOCs for 24 hours per day, 7 days per week. The two-year average levels of VOCs were used for residents' chronic exposure and were assumed to be representative of long-term exposure over eight years [15].

For cancer risks, our first step was to use the International Agency for Research on Cancer (IARC) classification system to group the measured VOCs. Then, we used the unit risk values proposed by the Cal-EPA for respective VOCs in the IARC Group 1 (carcinogenic to humans) or Group 2A (probably carcinogenic to humans) to calculate the potential cancer risks [16]. Again, two-year time-weighted average concentrations of VOCs were used to calculate cancer risks in specific target organs for all residents throughout the CTSP. We estimated a person's lifetime exposure on the assumption that a resident weighing 70 kg with a respiration rate of 20 m<sup>3</sup> per day lived on a sampling site for 24 h per day and 365 days per year for 70 years [16]. The lifetime cancer risk (LCR) for each VOC was calculated using Equation (3):

$$LCR_i = UR_i \times \bar{C}_i, \quad (3)$$

where  $LCR_i$  is the estimated risk from VOC  $i$  exposure,  $\bar{C}_i$  represents the two-year time-weighted average concentration of VOC  $i$  ( $\mu\text{g}/\text{m}^3$ ), and  $UR_i$  is the unit cancer risk ( $(\mu\text{g}/\text{m}^3)^{-1}$ ) for a given VOC.

### 2.6. Statistical Analysis

The Shapiro–Wilk test was first used to evaluate the normality of VOC concentration distribution for different environments (indoor or outdoor), measured seasons (compared with the spring), and monitored periods (2006 or 2007). The t-test was applied to compare the difference in specific VOCs and total VOCs between the indoor and outdoor measurements due to the concentration distributed

normally from the Shapiro–Wilk test (all  $p$ -values  $> 0.05$ ). The Wilcoxon rank-sum test was used to compare the seasonal difference in specific VOCs between 2006 and 2007. All the analyses were performed using the Statistical Analysis Software (SAS) standard package for Windows version 9.1 (SAS Institute Incorporation, Cary, NC, USA), and the significance level was set at 0.05.

### 3. Results

Table 1 shows the average concentrations of indoor and outdoor VOCs over two years. Ethanol (indoor:  $77.8 \pm 92.8 \mu\text{g}/\text{m}^3$ ; outdoor:  $36.8 \pm 29.6 \mu\text{g}/\text{m}^3$ ), toluene ( $67.0 \pm 36.7 \mu\text{g}/\text{m}^3$ ;  $56.9 \pm 19.0 \mu\text{g}/\text{m}^3$ ), m/p-xylene ( $50.8 \pm 66.1 \mu\text{g}/\text{m}^3$ ;  $21.2 \pm 20.3 \mu\text{g}/\text{m}^3$ ), and acetone ( $37.7 \pm 27.5 \mu\text{g}/\text{m}^3$ ;  $25.8 \pm 9.8 \mu\text{g}/\text{m}^3$ ) were identified as four dominant components both in the indoor and outdoor environments. Most of these VOCs were associated with traffic emissions except ethanol, acetone, IPA, cyclohexanone, and PGMEA, which were mainly detected in the optoelectronics plants [4,11,12]. The target compounds on the TO-15 method included benzene, toluene, ethylbenzene, m/p-xylene, o-xylene, styrene, methyl ethyl ketone (MEK), and methyl tertiary-butyl ether (MTBE). However, other VOCs of ethanol, acetone, ethyl acetate, IPA, cyclohexanone, and PGMEA were reviewed from the GC/MS data to examine the presence of compounds in the emission inventory. The total VOCs and 6 of 14 VOCs measured in the indoor sites were significantly higher than those measured in the outdoor sites, including benzene, toluene, ethylbenzene, m/p-xylene, o-xylene, and ethanol (all  $p$ -values  $< 0.05$ ). For the other eight VOCs, including styrene, acetone, MEK, ethyl acetate, IPA, MTBE, cyclohexanone, and PGMEA, there were no statistically significant differences between indoor and outdoor concentrations.

**Table 1.** Average concentrations ( $\mu\text{g}/\text{m}^3$ ) of indoor and outdoor volatile organic compounds (VOCs) over two years.

Chemicals	2006–2007 Outdoor	2006–2007 Indoor
	(Mean $\pm$ SD, N = 80) (Range)	(Mean $\pm$ SD, N = 72) (Range)
Benzene	4.9 $\pm$ 2.8 (2.3–10.1)	7.0 $\pm$ 4.1* (3.6–12.6)
Toluene	56.9 $\pm$ 19.0 (20.2–177.2)	67.0 $\pm$ 36.7* (26.4–148.4)
Ethylbenzene	7.8 $\pm$ 5.1 (4.2–15.7)	17.1 $\pm$ 22.4* (6.0–45.1)
m/p-Xylene	21.2 $\pm$ 20.3 (13.5–51.2)	50.8 $\pm$ 66.1* (16.1–138.5)
o-Xylene	6.6 $\pm$ 4.9 (4.3–11.3)	17.1 $\pm$ 23.4* (5.9–55.1)
Styrene	1.7 $\pm$ 1.3 (0.4–2.7)	2.5 $\pm$ 1.6 (0.7–6.0)
Acetone	25.8 $\pm$ 9.8 (16.9–34.9)	37.7 $\pm$ 27.5 (28.7–53.5)
Methyl ethyl ketone	14.9 $\pm$ 7.6 (7.8–36.4)	13.7 $\pm$ 6.8 (2.3–27.4)
Ethanol	36.8 $\pm$ 29.6 (9.0–88.4)	77.8 $\pm$ 92.8* (23.1–226.9)
Ethyl Acetate	10.7 $\pm$ 3.9 (4.0–27.2)	11.5 $\pm$ 6.8 (5.6–23.3)
Isopropyl alcohol	6.4 $\pm$ 3.1 (1.7–25.6)	5.2 $\pm$ 6.2 (2.8–12.6)
Others <sup>a</sup>	1.0 $\pm$ 1.7 (ND–4.8)	2.5 $\pm$ 3.3 (ND–11.4)
Total	213.6 $\pm$ 117.3 (148.6–465.7)	397.6 $\pm$ 303.2* (189.1–591.5)

<sup>a</sup> Including methyl tertiary-butyl ether (MTBE), cyclohexanone, and propylene glycol monomethyl ether acetate (PGMEA); \* Significant difference ( $p < 0.05$ ) for the t-test.

The seasonal variations of indoor VOC concentrations measured in 2006 and 2007 are shown in Table 2. Eight of fourteen VOCs measured in spring in 2007 were significantly higher than those measured in the same season in 2006, including benzene, toluene, ethylbenzene, m/p-xylene, o-xylene, MEK, ethyl acetate, and IPA (all  $p$ -values  $< 0.05$ ). In contrast, MEK monitored in summer and styrene monitored in autumn in 2007 were both significantly lower than those monitored in summer and autumn in 2006, respectively (both  $p$ -values  $< 0.05$ ).

**Table 2.** Seasonal variations of indoor VOC concentrations ( $\mu\text{g}/\text{m}^3$ ) in 2006 and 2007.

Chemicals	Spring		Summer		Autumn		Winter	
	2006 (N = 9)	2007 (N = 9)	2006 (N = 9)	2007 (N = 9)	2006 (N = 9)	2007 (N = 9)	2006 (N = 9)	2007 (N = 9)
Benzene	4.0 ± 1.2	12.6 ± 4.9*	9.8 ± 6.6	7.9 ± 4.3	3.6 ± 2.3	5.7 ± 3.8	6.0 ± 2.3	6.3 ± 7.0
Toluene	34.0 ± 8.8	148.4 ± 48.8*	95.9 ± 69.2	73.9 ± 16.9	32.4 ± 16.1	38.2 ± 20.6	26.4 ± 18.4	86.7 ± 94.4
Ethylbenzene	6.0 ± 2.2	29.0 ± 26.7*	18.2 ± 16.2	10.1 ± 4.8	7.3 ± 5.8	14.1 ± 24.2	6.7 ± 5.2	45.1 ± 94.2
m/p-Xylene	16.1 ± 7.0	75.8 ± 71.2*	53.8 ± 47.6	34.0 ± 16.3	18.0 ± 12.8	50.0 ± 86.4	20.4 ± 15.4	139 ± 272
o-Xylene	5.9 ± 2.3	15.5 ± 6.5*	24.3 ± 25.4	12.7 ± 7.0	6.1 ± 4.4	7.9 ± 11.5	9.0 ± 8.3	55.1 ± 121.6
Styrene	2.2 ± 0.6	2.5 ± 0.8	6.0 ± 2.3	3.6 ± 4.0	2.0 ± 1.1	0.7 ± 1.0*	1.9 ± 2.3	0.9 ± 1.0
Acetone	33.2 ± 34.8	28.7 ± 3.2	37.9 ± 34.2	43.9 ± 22.0	53.5 ± 65.4	34.2 ± 10.0	36.1 ± 30.2	34.1 ± 19.9
MEK	10.6 ± 1.7	27.4 ± 11.6*	15.1 ± 7.4	2.3 ± 3.2*	12.1 ± 2.2	22.6 ± 14.7	7.3 ± 4.1	12.3 ± 9.2
Ethanol	227 ± 364	94.9 ± 84.9	31.7 ± 21.4	73.1 ± 73.3	70.7 ± 89.6	53.1 ± 52.9	23.1 ± 22.0	49.2 ± 34.7
Ethyl Acetate	8.4 ± 6.6	23.3 ± 9.0*	12.4 ± 11.6	11.7 ± 7.5	8.4 ± 5.3	13.8 ± 5.6	5.6 ± 1.8	8.1 ± 6.9
Isopropyl alcohol	2.9 ± 2.2	5.9 ± 3.1*	2.8 ± 1.1	4.6 ± 6.4	4.9 ± 3.3	12.6 ± 26.6	3.3 ± 1.9	4.2 ± 4.9
Total	465 ± 350	554 ± 162	397 ± 289	355 ± 122	244 ± 95	386 ± 320	189 ± 84	492 ± 105

MEK, methyl ethyl ketone. \*: Significant difference ( $p < 0.05$ ) for the Wilcoxon rank-sum test.

Table 3 presents the residents' time activities in the indoor and outdoor environments during 2006–2007. The survey revealed that participants spent a mean of 16.6 h (69.2%) at home (i.e., living room, bedroom, and kitchen) and 7.4 h (30.8%) in the outdoor environment (including the workplace, commuting, and in other outside areas). The average times spent in the indoor and outdoor environments during 24 h over two years were applied to estimate the chronic exposure to specific VOCs for the health risk assessment.

**Table 3.** Average periods spent by residents in different indoor and outdoor environments over two years.

Environments	Hours (%)				
	Spring	Summer	Autumn	Winter	Average
Indoor					
Living room	6.0 (25)	5.6 (23.3)	8.0 (33.3)	8.9 (37.0)	7.1 (29.7)
Bedroom	7.2 (30)	8.3 (34.6)	7.9 (32.9)	8.0 (33.3)	7.9 (32.7)
Kitchen	1.9 (7.9)	1.7 (7.1)	1.4 (5.8)	1.4 (5.8)	1.6 (6.7)
Outdoor					
Workplace	4.0 (16.7)	4.0 (16.7)	4.0 (16.7)	2.7 (11.2)	3.7 (15.3)
Commute	0.6 (2.5)	0.4 (1.6)	0.5 (2.1)	0.2 (1.0)	0.4 (1.8)
Outside areas	4.3 (17.9)	4.0 (16.7)	2.2 (9.2)	2.8 (11.7)	3.3 (13.8)
Total	24 (100)	24 (100)	24 (100)	24 (100)	24 (100)

The estimated concentration and hazard index of chronic health effects (HIc) for residents exposed to six VOCs is shown in Table 4. Toluene was identified as the highest exposure among inhabitants living in the vicinity of the CTSP with a mean of 63.88 (range: 24.49–157.28)  $\mu\text{g}/\text{m}^3$ . Accordingly, the HIc due to toluene exposure was calculated as 0.213 (range: 0.082–0.524), which was associated with potential effects on development, nervous system, and alimentary system. While considering the additive effects of VOC exposure on the same target organ or system, the highest HIc was 0.397 (0.166–0.946) because of exposure to benzene, toluene, xylene, and styrene on the nervous system. However, either single or combined exposure of these six VOCs was estimated with the chronic non-carcinogenic risk less than unity.

Table 5 shows the potential cancer risk among residents living in the vicinity of the CTSP. Because only benzene was identified as a carcinogen on the hematopoietic system (i.e., leukemia) to humans (i.e., IARC group 1) and no other VOC was classified as group 1 or 2A according to the IARC system, the exposure level of benzene was conservatively applied for cancer risk assessment. Inhabitants were exposed to the benzene concentration of 6.10 (range: 3.20–11.83)  $\mu\text{g}/\text{m}^3$  during 2006–2007. The exposure data were further calculated with the lifetime cancer risk of  $1.8 \times 10^{-4}$  (range:  $9.3 \times 10^{-5}$ – $3.4 \times 10^{-4}$ ), which was higher than the general acceptable level of  $1.0 \times 10^{-6}$  for the public.

**Table 4.** Estimated levels ( $\mu\text{g}/\text{m}^3$ ) and hazard index of chronic health effects (HIC) for residents exposed to six VOCs in the vicinity of an optoelectronics industrial park.

Chemicals	Chronic REL ( $\mu\text{g}/\text{m}^3$ )	Estimated Levels	HIC	Target Organs or System
		Mean (range)	Mean (range)	
Benzene	60	6.10 (3.20–11.83)	0.102 (0.053–0.197)	Hematopoietic system; development; nervous system
Toluene	300	63.88 (24.49–157.28)	0.213 (0.082–0.524)	
Ethylbenzene	3000	14.23 (5.45–36.04)	0.005 (0.002–0.012)	Alimentary system (liver); kidney; development
Xylene	700	55.54 (20.71–153.18)	0.079 (0.030–0.219)	Nervous system; eye
Styrene	900	2.25 (0.61–4.98)	0.003 (0.001–0.006)	Nervous system
Isopropyl alcohol	7000	5.57 (2.46–16.61)	0.001 (<0.001–0.002)	Kidney
Benzene			0.102 (0.053–0.197)	Hematopoietic system
Benzene, toluene, xylene, and styrene			0.397 (0.166–0.946)	Nervous system
Toluene and ethylbenzene			0.218 (0.084–0.536)	Alimentary system
Benzene, toluene, and ethylbenzene			0.320 (0.137–0.733)	Development
Ethylbenzene and isopropyl alcohol			0.006 (0.002–0.014)	Kidney
Xylene			0.079 (0.030–0.219)	Eye

REL, reference exposure level.

**Table 5.** Estimated levels ( $\mu\text{g}/\text{m}^3$ ) and potential cancer risk for residents exposed to benzene in the vicinity of an optoelectronics industrial park.

Chemicals	Unit Risk ( $\mu\text{g}/\text{m}^3$ ) <sup>-1</sup>	IARC Group	Estimated Levels	Cancer Risk	Target Organs or System (Tumor Type)
			Mean (range)	Mean (range)	
Benzene	$2.9 \times 10^{-5}$	1	6.10 (3.20–11.83)	$1.8 \times 10^{-4}$ (9.3 $\times 10^{-5}$ – $3.4 \times 10^{-4}$ )	Hematopoietic system (leukemia)

IARC, International Agency for Research on Cancer.

## 4. Discussion

### 4.1. Indoor and Outdoor Exposure Assessment

We found that the four VOCs of ethanol, toluene, m/p-xylene, and acetone were dominant components in the indoor and outdoor environments for residents living near this optoelectronics industrial park. These VOCs were also identified as the major compounds reported in the ambient outdoor environment during 2005 [10], but were different from those of IPA, acetone, benzene, toluene, and PGMEA observed in a semiconductor industrial park [17] or those of ethylene, MEK, N, N-dimethylformamide, acrylonitrile, and toluene determined in a petrochemical industrial park [18]. The priority of indoor VOC levels was ethanol, toluene, m/p-xylene, and acetone, but the rank of outdoor ones changed to toluene, ethanol, acetone, and m/p-xylene in the present study. Ethanol and acetone were reported as the major compounds measured in a TFT-LCD company [4], and the toluene and m/p-xylene may be from the sources of traffic and industrial activities [10,19].

Benzene, toluene, ethylbenzene, and xylene (i.e., m/p-xylene and o-xylene) are most common compounds of VOCs emitted from traffic [19], but partial levels may be released from the TFT-LCD manufacturing processes in this park. One previous study reported the detected level of benzene in the development, etching, photoresist stripping, rubbing and polyimide printing, spacer sealant, and hot press and cell attachment [11]. Both toluene and xylene were detected in the workplace of TFT-LCD companies [4,11]. Ethylbenzene was not identified in previous studies. In comparison with the major VOCs of ethanol and acetone measured in the manufacturing processes, the concentration ratio ranged from 0.4% (toluene/ethanol) [4] to 5% (toluene/acetone) [11].

We did not detect TMAH in either indoor or outdoor environments. TMAH is a widely utilized etchant in the optoelectronic manufacturing processes [13]. Dermal exposure to TMAH may result in

alkalinity-related chemical burns, respiratory failure, and/or sudden death in humans [20,21]. Because of its ionic nature and low volatility, it is almost impossible to detect TMAH in ambient air.

In addition, the measured indoor levels of VOCs were obviously higher than those measured in the outdoor environment. These results were consistent with findings reported in previous studies [22,23]. Our questionnaire survey found that 39% of participants burned incense at home, 22% of inhabitants used fragrance inside, and 8% of residents had painted within the last one month. This indicated possible VOC sources emitted from the indoor environment. As only 17% of participants admitted using air conditioners during the sampling period, the indoor levels of VOCs were partially contributed to the outdoor sources in this study.

We also observed the highest concentration of indoor total VOCs in spring compared with those measured in other seasons in both 2006 ( $465 \pm 350 \mu\text{g}/\text{m}^3$ ) and 2007 ( $554 \pm 162 \mu\text{g}/\text{m}^3$ ). This is because 97% of subjects reported that they usually open windows for ventilation in the spring, which produces the highest indoor level of total VOCs in the present study.

#### 4.2. Health Risk Assessment

We did not observe any chronic hazard indices of single or mixed VOCs greater than unity, which implied that no non-carcinogenic health effects were expected. The same results were reported in people living in the vicinity of the largest chemical production site in the Mediterranean area [24] and among workers in the rubber footwear industries in China [25]. In contrast, one previous study in southern Taiwan estimated the excess non-cancer risk among petrochemical workers as a result of exposure to acrylonitrile (respiratory system), 1,3-butadiene (reproductive system), hydrogen cyanide (nervous system, endocrine system, and cardiovascular system), and *n,n*-dimethylformamide (alimentary system and respiratory system) [18]. These differences revealed the variations in emitted sources and concentrations of VOCs between different industrial areas.

We observed a cancer risk due to benzene of greater than the acceptable level of  $10^{-6}$  for lifetime exposure. The mean cancer risk of  $1.8 \times 10^{-4}$  higher than  $1.0 \times 10^{-4}$  was categorized as a definite risk [26]. The results were similar to other findings in different industrial areas, including oil-burning power and steel plants in Japan [23], the petrochemical complex in southern Taiwan [18], the largest chemical site in southern Europe and the Mediterranean region [24], and the rubber footwear industries in China [25]. All of these studies identified benzene exposure to be one of the highest cancer risks for workers and residents living in the vicinity of the industry. We recommend long-term monitoring of benzene levels and actions taken to reduce the emitted sources for public health.

This study presented the preliminary results conducted in 2006–2007 for residents living in the vicinity of the optoelectronics industrial park. The two-year average levels of indoor and outdoor VOCs, as well as the survey of residents' time activities, were used to estimate the potential non-carcinogenic and carcinogenic risks for inhabitants. On the basis of these findings, a follow-up survey was performed in 2019 to measure the indoor and outdoor levels of VOCs. We will share the comparison of results between 2006 and 2019 with readers in the future.

#### 4.3. Strengths and Limitations

This study had the strength of combining the indoor and outdoor measurements of VOCs with time-activity patterns over two years for residents living near this optoelectronics park. Such detailed exposure data provided more precise estimations of potential non-carcinogenic and carcinogenic risks among inhabitants. However, some limitations still have to be mentioned. First, we only performed a 24-h sampling each season and used four-day results to present the annual levels. These results could not be completely representative for seasonal variations and could have been accidental. Greater frequencies of VOCs sampling (i.e., per week or per month) are preferable, but not practical. A fixed monitoring site of VOCs can overcome the limitation of inadequate sampling frequency, but no continuous monitoring stations have been set up around this park. Second, indoor sampling of VOCs in the living room instead of personal sampling restricted the investigation for exposure in different

microenvironments (e.g., bedroom, kitchen, and study). This drawback may reduce the accuracy of the estimated exposure. Third, some standard deviations of indoor VOCs (such as ethylbenzene, m/p-xylene, o-xylene, ethanol, and IPA) exceeded the annual average levels, particularly in winter 2007, indicating greater variations of indoor exposure in the microenvironment. Future studies have to extend the monitoring durations (i.e., one week or more in the house) to decrease the exposure variation. Fourth, a major limitation of this study is the lack of upwind monitoring and comparison of upwind and downwind data around the two clusters of sampling sites around this industrial park to determine the sources of VOCs. Fifth, traffic conditions around the sampling sites were not measured, even though the compounds of benzene, toluene, ethylbenzene, and xylene can be traffic-related. Sixth, the two-year monitoring mean, but not an eight-year (i.e., 12% of human lifetime) [15,16] average was applied to estimate the chronic non-cancer and cancer risks. Because this optoelectronics park has been in operation for just over three years (since 2003), the emitted amount of VOCs may be less than that after a longer running period with growing production over time. This might underestimate the residents' chronic exposure. Finally, some default assumptions from risk assessment, such as 20 m<sup>3</sup> inhaled air per day, daily exposure to VOC levels on 365 days per year, occupation of the current address over 70 years, and residents with the average weight of 70 kilograms, may generate uncertainty in cancer risk evaluation. A more detailed survey that includes this information can improve the accuracy of our estimated results.

## 5. Conclusions

This study revealed that residents living near this optoelectronics park had higher indoor exposure to VOCs than outdoor exposure, primarily from ethanol, toluene, m/p-xylene, and acetone. The estimated cancer risk for inhabitants was above the general acceptable level as a result of benzene exposure. Long-term monitoring of VOCs and reduction of benzene exposure in residents should be performed to monitor the human exposure to air toxins and ensure the protection of public health.

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**Conflicts of Interest:** The authors declare no conflict of interest.

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