



Long-term Multiple Chemical Exposure Assessment for a Thin Film Transistor Liquid Crystal Display (TFT-LCD) Industry

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ABSTRACT

A surrogate approach was deployed for assessing long-term exposures of multiple chemicals at 8 selected working areas of 3 manufacturing processes located at a clean room of a thin film transistor liquid crystal display (TFT-LCD) industry. For each selected area, 6 to 12 portable photoionization detector (PID) were placed uniformly in its workplace to measure its total VOCs concentrations (C_{T-VOCs}) for 6 randomly selected workshifts. Simultaneously, one canister was placed beside one of these portable PIDs, and the collected air sample was analyzed for individual concentration (C_{VOCi}) of 107 VOCs. Predictive models were established by relating the C_{T-VOCs} to C_{VOCi} of each individual compound via simple regression analysis. The established predictive models were employed to construct a year-long C_{VOCi} databank based on the measured year-long C_{T-VOC} for each selected area using the same portable PID. The ethanol (381 ppb–2,480 ppb), acetone (123 ppb–624 ppb) and propylene glycol monomethyl ether acetate 29 (PGMEA; 14.4 ppb–2,241 ppb) dominated in all selected areas, and all measured C_{VOCi} were much lower than their permissible exposure limits. Predictive models obtained from simple linear regression analyses were found with an $R^2 > 0.453$ indicating that C_{T-VOCs} were adequate for predicting C_{VOCi} . The predicted year-long C_{VOCi} reveals that long-term total multiple chemical exposures of all selected areas fall to the range 0.10%–20% of the permissible exposure level. Using the C_{T-VOCs} as a surrogate for the routine checking purpose, the present study yielded allowable C_{T-VOCs} fall to the ranges of 49.1 ppm–577 ppm. Considering the approach used in the present study requires less cost and manpower, it would be applicable to similar industries for conducting long-term multiple chemical exposure assessments in the future.

Keywords: Multiple chemical exposures; Long-term exposure assessment; Risk assessment; Clean room.

INTRODUCTION

For high-tech industries, many studies have shown their exposure concentrations of an individual compound (such as ethanol, acetone, butyl acetate, isopropyl alcohol (IPA), and propylene glycol monomethyl ether acetate (PGMEA)) could be much lower than that of traditional industry, and particularly lower than their time-weighted-average permissible exposure concentrations (PEL-TWAs) (Zappe,

1995; Chuah *et al.*, 2000; Wu *et al.*, 2004; Chang *et al.*, 2010a; Park *et al.*, 2011; Cheng *et al.*, 2016; Chou *et al.*, 2016; Guerra *et al.*, 2017). Apparently, chemical exposures in the high-tech industry might not cause significant health risk posed on workers. However, recent epidemiological studies conducted in a photolithography industry have shown that long-term exposure to ethanol and acetone might result in workers with irregular menstrual cycles and endocrine disruption (Hsieh *et al.*, 2005; Lin *et al.*, 2013a). In a liquid crystal display (LCD) manufacturing factory, long-term exposures to multiple chemicals of low levels might lead to premenopausal female workers with modest changes in their reproductive hormone urinary concentrations (Lin *et al.*, 2013b; Chan *et al.*, 2017). A slightly increase in kidney dysfunction risk was identified for workers in a TFT-LCD

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industry (simultaneously exposed to IPA, PGMEA, and propylene glycol monomethyl ether (PGME)) with their total exposure levels ≥ 50 ppb for 3–5 years (Chang *et al.*, 2010b).

Considering the exposure concentration of a chemical might be varied greatly over time in the high-tech industry, conducting long-term exposure assessment has become a crucial issue, especially for those chemicals with chronic health effects. But it is known that huge manpower is usually required for conducting long term exposure assessment. As a result, the long-term exposure databank is only sparsely found in the industry. Since multiple chemicals are simultaneously used in the high-tech industry, the total health effect can be assessed only if multiple chemical exposure assessments were conducted. Yet, it is true that some current available sampling methods can be used to collect multiple chemicals in one sample and do the analysis simultaneously (e.g., the NIOSH method 2549; using thermal desorption tube/gas chromatography, mass spectrometry for analyses (NIOSH, 1996)). These methods might be also not widely adopted in the field because of the constraints of the cost associated with chemical analyses. Therefore, developing approaches which require less manpower and lower cost has become an important issue in the industrial hygiene field.

To solve the above problems, the method associated with measuring surrogate concentrations through the use of a direct reading instrument has been adopted by industrial hygienists recently (Hewett *et al.*, 2006; Chen *et al.*, 2012; Hsu *et al.*, 2012). The applicability of the above method can be assessed if the measured surrogate concentrations could effectively predict the individual chemical exposure level of interest. For example, a strong correlation has been found between the 2-butoxyethanol concentrations measured using sorbent tubes and the concentrations of total volatile organic carbons (C_{T-VOCs}) measured using a portable photoionization detector (PID) (Hewett *et al.*, 2006; Bello *et al.*, 2013). In addition, the above instrument has been recognized with advantages in its operability and low operation cost. Therefore, its long-term measured results, after being converted to chemical concentrations of interest, could be used for establishing a long-term exposure data. Considering a large amount of samples are required for establishing a long-term exposure databank, using the hand-held VOC monitor is obviously an alternative from both the cost and applicability aspects.

In the present study, C_{T-VOCs} was adopted as a surrogate, together with the monitoring of individual compounds of interest, a long-term multiple chemical exposure assessment was conducted in a TFT-LCD industry. Considering the proposed approach requires less cost and manpower, it would provide a possible solution for many other industries for conducting long-term multiple chemical exposure assessments in the future.

MATERIAL AND METHODS

Sampling Strategy

In the present study, three manufacturing processes in a TFT-LCD manufacturing industry were selected for conducting field samplings, including the Array process

(mainly for producing TFT-plates), CF process (mainly for manufacturing color filter-plates (CF-plates)), and Cell process (mainly for filling liquid crystal to assembly a TFT plate and a CF into a TFT-LCD plate). The above three processes were further divided into eight working areas, including four areas in the Array process (i.e., CVD, Dry, Wet, and Photo), three areas in the CF process (i.e., ITO, BM/R, and G/B), and one area in the Cell process (i.e., PI). A portable photoionization detector (PID) (ppbRAE 3000, PGM-7340, RAE System Inc., San Jose, CA, USA; range of detection = 1 ppb–10,000 ppm) was used to measure the concentrations of the total VOCs (C_{T-VOCs}). In the present study, six to twelve hand-held VOC monitors were placed uniformly in each selected working area to measure its total VOC concentrations (C_{T-VOCs}) for six workshifts (including three day-shifts (from 09:00 AM to 11:00 AM) and three night-shifts (22:00 PM to 24:00 PM) in three days randomly selected during one week). Simultaneously, one 6-L silonite canister (flowrate = 40 mL min⁻¹) was placed beside one hand-held VOC monitor for collecting an air sample, and the collected sample was analyzed for the concentrations of each individual VOCs (C_{VOCi}). The selection of six workshift samplings was because the sample size would be a reasonable approximation of an exposure distribution (Mulhausen *et al.*, 2006). Sampling probes of both hand-held VOC monitor and canister were placed at the height ~1.5 m above the floor to simulate workers' breathing zone exposures. The above sampling results were used to examine the relationship between C_{T-VOCs} and C_{VOCi} for samples collected from each of the eight selected areas.

In addition, the hand-held VOC monitor was also used for conducting samplings in each selected area one or two times per day for one year. As a result, more than 365 samples were obtained for each investigated area. The obtained C_{T-VOCs} were further converted to C_{VOCi} to establish the year-long C_{VOCi} exposure databank.

Analyzing for Volatile Organic Compounds (VOCs)

For each collected canister sample, analyzed compounds were determined partly based on the safety data sheets (SDS) provided by the safety and health personnel of the industry, and some were by reference to researches related to this industry (Hsieh *et al.*, 2005; Park *et al.*, 2011; Lin *et al.*, 2013a, b; Chang *et al.*, 2010b; Yang *et al.*, 2016). As a result, a total of 107 VOCs were included for sample analyses. Analyses of VOCs were performed in accordance with the Compendium method PAMS (Photochemical Assessment Monitoring System) and TO-15 (U.S. EPA, 1999). The VOCs collected in the canister were concentrated in a cryogenic concentrator (Entech 7100A, Entech Instruments Inc.) and then injected into a gas chromatography/mass spectrometry (GC/MS) (HP6980 GC & HP5973 MSD, Agilent Technologies Inc., Santa Clara, CA, USA) with a J&B DB-5MS capillary column (60 m \times 0.25 mm \times 1.0 μ m, J&B Scientific, Folsom, CA, USA). The detection limits for all analyzed VOCs fell within the range of 0.0002 ppb (n-Dodecane) –3.80 ppb (Methanol), and the relative standard deviation for all VOCs were < 8.81%.

Data Analyses

Since there is a priori knowledge that if both hand-held VOC monitor and canister were placed in a hypothetical environment without the existence of any organic solvent, there is no doubt that both C_{T-VOCs} and C_{VOCi} would become zero. Therefore, a simple linear regression model with the regression line forced through the origin was employed to examine the relationship between the C_{VOCi} and C_{T-VOCs} based on the suggestion of Neter *et al.* (1985):

$$C_{VOCi} = \alpha C_{T-VOCs} \quad (1)$$

Considering both C_{VOCi} and C_{T-VOCs} were collected simultaneously from the same site, a similar concentration distribution can be expected for both measured C_{VOCi} and C_{T-VOCs} . We selected six (including 5th, 15th, 25th, 50th, 75th, and 95th) percentile concentrations of the two distributions to conduct the above simple linear regression analyses. If a good correlation could be found for a given studied area, the above regression equation could be used to further convert its year-long C_{T-VOCs} to establish a long-term C_{VOCi} exposure databank.

Considering workers in these eight selected areas were exposed to multiple chemicals associated with their permissible exposure limits (PELs) set on different criteria. Though different methodologies have been developed for assessing multiple chemical exposures (Haddad *et al.*, 2001; Meek *et al.*, 2011; Sargiannis *et al.*, 2012), no universally applicable methodology does yet exist because of the complexities in their exposure routes, target organs, and pharmacokinetic interactions among chemicals present as mixtures. For the practical use in the field, a pragmatic approach has been proposed by U.S. EPA (U.S. EPA, 1986). The approach involves the summation of excess risk attributed to each mixture constituent. The approach neither considers the information on target tissue dose of mixture nor accounts for potential interactions occurring at the pharmacokinetic level. The result obtained from the approach can be considered as the "worse-case" which would be conservative for assessing workers' exposures and the corresponding health risks. For a multiple chemical exposure scenario, its exposure level can be determined by the following:

$$\text{Total Exposure} = \frac{C_{VOC_1}}{PEL - TWA_1} + \frac{C_{VOC_2}}{PEL - TWA_2} + \dots + \frac{C_{VOC_n}}{PEL - TWA_n} \quad (2)$$

where, C_{VOCi} and $PEL - TWA_i$ represent the level of the i^{th} volatile organic compound and its corresponding $PEL - TWA$, respectively. If a total exposure was above the unity indicating that the multiple chemical exposure scenario was unacceptable.

RESULTS AND DISCUSSION

Concentrations of Each Individual VOC (C_{VOCi}) in the

Eight Selected Areas

Table 1 shows the concentrations of C_{VOCi} obtained from the eight studied areas using canisters for collecting air samples during one week. In total, thirty-two compounds were detected from the eight selected areas, and seventeen to twenty-eight compounds were detected for each area. Among all detected compounds, one compound belongs to the IARC group 2a (i.e., methylene chloride) and another five compounds belong to IARC group 2b (i.e., naphthalene, isopropyl benzene, ethyl benzene, styrene, and MIBK), respectively. The eight compounds of the ethanol, acetone, and PGMEA, 2-propanol, 1,2,3-trimethylbenzene, 1,3,5-trimethylbenzene, ethyl acetate, and 1,2,4-trimethylbenzene have been confirmed as the raw materials used in manufacturing processes after examining the safety data sheets (SDSs) of these areas. In addition, the five compounds of the o-xylene, p-xylene, m-xylene, toluene, and MTBE could be associated with traffic emissions (NIOSH, 1996; Chiu *et al.*, 2005; Hsieh *et al.*, 2006). The two compounds of the toluene and phenol have been identified as the byproducts associated with the thermal decomposition of the photoresist (OEHHA Toxicity Criteria Database, 2015). Previous study also reported that if the local scrubber wasn't well maintained in TFT-LCD manufacturing processes, toxic chlorinated byproducts, such as trichlorofluoromethane, would be found in the process areas (Park *et al.*, 2011). On the other hand, there are at least sixteen VOCs detected from the studied areas which might belong to unknown byproducts or unreleased raw materials used in the manufacturing processes. The above results further confirm that the exposure scenarios of workers in the clean room of the TFT-LCD industry are very complicated, which is consistent with the conclusion made by one previous study (Park *et al.*, 2011).

All C_{VOCi} obtained from these eight studied areas were well below the corresponding PELs regulated in Taiwan. The above results were consistent with those of other studies (Chang *et al.*, 2010a; Park *et al.*, 2011; Shie *et al.*, 2011). Among these detected compounds, the ethanol (381 ppb–2,480 ppb), acetone (123 ppb–624 ppb), and PGMEA (14.4 ppb–2,241 ppb) were compounds dominated in all studied areas. The above findings are also very similar to those found in previous studies. For example, Chang *et al.* (2010a) found that the ethanol (656.8 ppb–2,594 ppb), acetone (343.3 ppb–2066 ppb), isopropyl alcohol (1.7 ppb–388.5 ppb), and PGMEA (0.9 ppb–205.9 ppb) were the four dominant compounds in a total of ten VOCs identified in a TFT-LCD factory. Wu *et al.* (2004) found the concentrations of PGMEA (252.7 ppb), butyl acetate (237.0 ppb), and acetone (75.9 ppb) in the workplace atmosphere of LCD fabrication facilities. Park *et al.* (2011) found a maximum exposure concentration of PGMEA of 87.0 ppb in a wafer fabrication industry facility. All these reports support that the concentration of individual compound in the hi-tech industry is low as in comparison with its $PEL - TWA$.

Concentrations of total VOCs (C_{T-VOCs}) in the Eight Selected Areas

Table 2 shows the concentrations of total VOCs (C_{T-VOCs}) obtained from the selected eight areas using a portable PID

Table 1. Concentrations of each individual VOC (C_{VOC} ; ppb) (Mean \pm SD) obtained from the eight studied areas using canisters during one week (n = 6).

CAS NO.	Chemical	Array				CF				Cell
		CVD	Dry	Wet	Photo	ITO	BM/R	G/B	PI	
00064-17-5	Ethanol	777 \pm 442	741 \pm 614	442 \pm 295	463 \pm 447	381 \pm 227	626 \pm 521	516 \pm 192	2480 \pm 2107	
00067-63-0	2-Propanol	-	12.1 \pm 5.82	15.2 \pm 1.99	22.1 \pm 4.60	5.73 \pm 2.54	5.16 \pm 1.06	-	31.1 \pm 10.7	
00067-64-1	Acetone	221 \pm 51.4	155 \pm 115	138 \pm 93.4	123 \pm 95.4	305 \pm 182	544 \pm 353	624 \pm 371	530 \pm 274	
00075-09-2	Methylene Chloride*	-	19.7 \pm 9.4	32.2 \pm 11.2	45.2 \pm 15.3	22.7 \pm 10.2	18.4 \pm 7.70	35.8 \pm 13.6	10.7 \pm 3.45	
00075-69-4	Trichlorofluoromethane	-	-	-	-	1.92 \pm 0.86	1.92 \pm 0.41	-	-	
00076-13-1	1,1,2-Trichlorotrifluoroethane	-	6.57 \pm 3.13	8.78 \pm 3.05	-	-	-	-	-	
00078-78-4	Isopentane	-	-	-	6.37 \pm 2.74	-	10.4 \pm 2.14	-	-	
00078-93-3	Butanone	-	10.1 \pm 4.83	10.5 \pm 3.59	11.3 \pm 3.81	4.99 \pm 2.25	-	-	10.9 \pm 3.47	
00091-20-3	Naphthalene**	-	-	-	6.49 \pm 1.88	-	-	-	-	
00095-47-6	o-Xylene	1.38 \pm 0.09	1.27 \pm 0.24	1.30 \pm 0.27	1.36 \pm 0.28	0.58 \pm 0.04	0.59 \pm 0.05	0.64 \pm 0.15	0.84 \pm 0.19	
00095-63-6	1,2,3-Trimethylbenzene	0.67 \pm 0.09	0.56 \pm 0.21	0.54 \pm 0.29	0.62 \pm 0.18	0.22 \pm 0.02	0.35 \pm 0.08	0.38 \pm 0.08	0.26 \pm 0.03	
00098-82-8	Isopropylbenzene**	0.32 \pm 0.07	0.29 \pm 0.06	0.29 \pm 0.06	0.28 \pm 0.07	0.19 \pm 0.01	0.19 \pm 0.01	0.19 \pm 0.01	-	
00100-41-4	Ethyl Benzene**	2.65 \pm 0.64	2.39 \pm 1.25	2.53 \pm 1.28	2.07 \pm 1.19	0.68 \pm 0.30	0.72 \pm 0.15	0.88 \pm 0.35	0.94 \pm 0.20	
00100-42-5	Styrene**	-	0.27 \pm 0.06	0.27 \pm 0.09	0.27 \pm 0.08	0.16 \pm 0.07	0.16 \pm 0.03	0.15 \pm 0.05	0.23 \pm 0.05	
00103-65-1	n-Propylbenzene	0.43 \pm 0.09	0.39 \pm 0.07	0.40 \pm 0.08	0.39 \pm 0.08	0.33 \pm 0.11	-	0.32 \pm 0.11	0.34 \pm 0.08	
00106-42-3	p-Xylene	1.40 \pm 0.42	1.59 \pm 1.07	1.25 \pm 0.50	1.25 \pm 0.48	0.54 \pm 0.24	0.55 \pm 0.11	0.59 \pm 0.13	0.77 \pm 0.13	
00107-83-5	2-Methylpentane	-	-	-	-	-	0.82 \pm 0.17	-	-	
00108-10-1	MIBK**	-	8.76 \pm 4.17	9.69 \pm 3.31	9.67 \pm 3.32	4.76 \pm 0.09	4.74 \pm 0.98	4.23 \pm 1.23	9.47 \pm 3.02	
00108-38-3	m-Xylene	2.77 \pm 0.74	2.37 \pm 1.22	2.64 \pm 1.45	2.71 \pm 1.55	0.70 \pm 0.31	0.74 \pm 0.15	0.84 \pm 0.14	0.99 \pm 0.19	
00108-65-6	PGMEA	21.9 \pm 10.6	68.6 \pm 48.1	14.4 \pm 9.47	2241 \pm 810	43.1 \pm 16.5	38.9 \pm 27.9	741 \pm 738	777 \pm 653	
00108-67-8	1,3,5-Trimethylbenzene	0.34 \pm 0.02	0.33 \pm 0.005	0.33 \pm 0.02	0.32 \pm 0.03	0.22 \pm 0.06	0.19 \pm 0.05	0.24 \pm 0.09	0.18 \pm 0.03	
00108-88-3	Toluene	7.74 \pm 5.45	2.48 \pm 1.23	11.9 \pm 10.3	4.39 \pm 3.52	4.72 \pm 3.83	4.78 \pm 4.75	2.73 \pm 2.18	6.39 \pm 8.20	
00110-54-3	n-Hexane	-	1.71 \pm 0.81	2.06 \pm 0.88	7.60 \pm 2.25	4.56 \pm 1.63	4.35 \pm 2.41	4.63 \pm 1.76	8.44 \pm 0.80	
00111-84-2	n-Nonane	-	-	0.14 \pm 0.05	-	-	-	-	0.24 \pm 0.09	
00112-40-3	n-Dodecane	0.50 \pm 0.11	0.40 \pm 0.13	0.54 \pm 0.18	0.35 \pm 0.09	0.12 \pm 0.02	0.12 \pm 0.03	0.12 \pm 0.05	0.63 \pm 0.22	
00141-78-6	Ethyl Acetate	-	9.49 \pm 4.52	10.1 \pm 3.46	20.4 \pm 13.4	4.88 \pm 2.15	7.96 \pm 1.64	-	10.5 \pm 3.37	
00526-73-8	1,2,4-Trimethylbenzene	0.36 \pm 0.07	0.33 \pm 0.05	0.32 \pm 0.05	0.32 \pm 0.06	0.40 \pm 0.12	0.21 \pm 0.01	0.24 \pm 0.07	0.23 \pm 0.03	
00611-14-3	o-Ethyltoluene	0.33 \pm 0.03	0.31 \pm 0.02	0.32 \pm 0.02	0.32 \pm 0.05	0.18 \pm 0.08	0.31 \pm 0.06	0.35 \pm 0.13	0.31 \pm 0.07	
00620-14-4	m-Ethyltoluene	0.49 \pm 0.02	0.45 \pm 0.06	0.46 \pm 0.08	0.46 \pm 0.09	0.34 \pm 0.09	0.31 \pm 0.07	0.33 \pm 0.08	0.27 \pm 0.07	
00622-96-8	p-Ethyltoluene	0.46 \pm 0.11	0.41 \pm 0.09	0.40 \pm 0.09	0.41 \pm 0.09	0.34 \pm 0.08	0.31 \pm 0.08	0.33 \pm 0.09	0.27 \pm 0.08	
01120-21-4	n-Undecane	-	-	0.17 \pm 0.06	-	-	-	-	0.18 \pm 0.07	
01634-04-4	MTBE	-	12.4 \pm 4.66	15.0 \pm 5.21	12.9 \pm 2.91	-	11.5 \pm 2.61	-	14.7 \pm 4.69	

PGMEA: Propylene glycol monomethyl ether acetate; MIBK: 4-Methyl-2-Pentanone; MTBE: Methyl-tert-Butylether.

*: IARC group 2A; **: IARC group 2B.

"-": Non-detected.

during one week. In sequence, the levels of C_{T-VOCs} were Photo (3139 ppb) > Cell/PI (3046 ppb) > Wet (2312 ppb) > CVD (1872 ppb) > Dry (1810 ppb) > G/B (1339 ppb) > BM/R (1265 ppb) > ITO (1195 ppb) (ANOVA, $p < 0.01$). The highest C_{T-VOCs} in Photo was ~2.6 times in magnitude higher than that of ITO (the lowest).

Table 3 shows the year-long C_{T-VOCs} of the eight studied areas. The mean levels of C_{T-VOCs} , as shown in sequence, were Photo (5227 ppb) > Wet (3780 ppb) > Cell/PI (3633 ppb) > Dry (2795 ppb) > CVD (2789 ppb) > BM/R (2494 ppb) > G/B (2301 ppb) > ITO (1616 ppb). It also should be noted that the year-long C_{T-VOCs} levels were consistently higher (1.12–1.97 folds) than that of one-week measured C_{T-VOCs} levels (Table 3). The above significant differences between both C_{T-VOCs} levels could be due to variations in the productivity (or amount of raw material used) over time. In addition, it should be noted that the coefficient of variation (CV; = SD/mean) estimated for the eight selected areas based on one-week C_{T-VOCs} (range = 1.78%–9.71%; Table 2) were much less than the corresponding values obtained from the year-long C_{T-VOCs} (range = 34.8%–69.6%; Table 3). The above results further confirm the above inference, since the variation in the productivity within one-week should be much less than that of the year-long. Therefore, it is concluded that cross-sectional monitoring (i.e., one-week sampling) results were inadequate for conducting long-term exposure assessment.

Estimation of Long-Term C_{VOCi} for the Eight Selected Areas

In the present study, simple linear regression analyses (see Eq. (1)) were employed to relate C_{VOCi} to C_{T-VOCs} based on the one-week measured data. Resultant regression

models were applied to the measured year-long C_{T-VOCs} for predicting the year-long C_{VOCi} . Results show that all resultant regression coefficients (i.e., the α value in Eq. (1)) were found with positive values and were significant statistically ($p < 0.01$). Table 4 shows resultant regression models of the measured year-long C_{T-VOCs} to predict the year-long C_{VOCi} , the obtained R^2 for the eight selected areas fall to the range 0.826–0.997 for CVD, 0.603–0.998 for Dry, 0.717–0.997 for Wet, 0.472–0.991 for Photo, 0.661–0.998 for ITO, 0.854–0.992 for BM/R, 0.819–0.997 for G/B, and 0.453–0.990 for Cell/PI, respectively. Among them, the two lowest R^2 were obtained for toluene in Photo ($R^2 = 0.472$) and n-hexane in Cell/PI ($R^2 = 0.453$). Since the above two compounds were mainly transported from the outside ambient air, the low R^2 could be affected by their intrinsic concentration variations in the ambient air. Nevertheless, the high R^2 obtained from the present study indicates that the proposed surrogate (C_{T-VOCs}) was adequate for predicting C_{VOCi} for the eight selected areas.

By applying all resultant predictive models, the measured year-long C_{VOCi} were converted to the year-long C_{T-VOCs} to for the eight selected areas (Table 5). Since the levels of the year-long C_{T-VOCs} (Table 3) were higher than that of one-week C_{T-VOCs} for all studied areas (Table 2), it was expected that the estimated year-long C_{VOCi} (Table 6) should be higher than the corresponding one-week measured C_{VOCi} (Table 1). However, some year-long C_{VOCi} levels were found to be lower than that of one-week measured C_{VOCi} (such as the 1,2,3-Trimethylbenzene in the four areas of the CVD, Dry, Wet, and Photo; n-hexane, ethyl acetate, and MTBE in Dry, 1,2,4-trimethylbenzene in Photo; toluene, n-hexane, n-nonane, ethyl acetate; MTBE in Cell/PI). Here, it should be noted that there are intrinsic differences

Table 2. Concentrations of total VOCs (C_{T-VOCs}) obtained from the eight studied areas using the hand-held VOC monitors during one week (ppb) (n = 6).

Process	Area	Mean	SD	Max	Min	CV
Array	CVD	1872	33.4	2789	1391	1.78%
	Dry	1810	23.4	2926	1302	1.29%
	Wet	2312	142	3422	1555	6.14%
	Photo	3139	289	4774	1598	9.21%
CF	ITO	1195	59.3	1993	704	4.96%
	BM/R	1265	100	2939	747	7.91%
	G/B	1339	114	3201	785	8.51%
Cell	PI	3046	176	5506	1926	5.78%

Table 3. Year-long concentrations of total VOCs (C_{T-VOCs} ; ppb) obtained from the eight studied areas using the hand-held VOC monitors.

Process	Area	Mean	SD	Max	Min	CV
Array (n = 1,729)	CVD	2789	1553	14957	866	55.7%
	Dry	2795	1673	15907	723	59.9%
	Wet	3780	2629	24383	524	69.6%
	Photo	5227	3469	32410	931	66.4%
CF (n = 1,216)	ITO	1616	563	4622	542	34.8%
	BM/R	2494	1367	9810	43	54.8%
	G/B	2301	1104	8194	194	47.9%
Cell (n = 384)	PI	3633	1533	7768	1118	42.2%

Table 4. Resultant regression models of the measured year-long C_{T-VOCs} to predict the year-long C_{VOC1} for the eight studied areas.

CAS NO.	Chemical	ITO						CF						Cell					
		R ²		p-value		α		BM/R		p-value		α		G/B		PI			
		α	R ²	p-value	α	R ²	p-value	α	R ²	p-value	α	R ²	p-value	α	R ²	p-value			
00064-17-5	Ethanol	3.22×10^{-1}	0.956	7.27×10^{-4}	5.31×10^{-1}	0.860	7.73×10^{-3}	3.64×10^{-1}	0.997	2.85×10^{-6}	8.58×10^{-1}	0.863	3.59×10^{-5}						
00067-63-0	2-Propanol	4.88×10^{-3}	0.979	1.64×10^{-4}	3.87×10^{-3}	0.980	1.46×10^{-4}	-	-	-	1.02×10^{-2}	0.783	2.98×10^{-4}						
00067-64-1	Acetone	2.63×10^{-1}	0.961	5.93×10^{-4}	4.39×10^{-1}	0.964	4.79×10^{-4}	4.67×10^{-1}	0.969	3.73×10^{-4}	1.79×10^{-1}	0.906	6.54×10^{-6}						
00075-09-2	Methylene Chloride*	1.93×10^{-2}	0.979	1.64×10^{-4}	1.34×10^{-2}	0.980	1.46×10^{-4}	2.55×10^{-2}	0.992	2.60×10^{-5}	3.56×10^{-3}	0.989	4.70×10^{-5}						
00075-69-4	Trichlorofluoromethane	1.63×10^{-3}	0.979	1.64×10^{-4}	1.40×10^{-3}	0.980	1.46×10^{-4}	-	-	-	-	-	-						
00076-13-1	1,1,2-Trichlorotrifluoroethane	-	-	-	-	-	-	-	-	-	-	-	-						
00078-78-4	Isopentane	-	-	-	7.61×10^{-3}	0.980	1.46×10^{-4}	-	-	-	-	-	-						
00078-93-3	Methyl ethyl ketone	4.25×10^{-3}	0.979	1.64×10^{-4}	-	-	-	-	-	-	3.64×10^{-3}	0.989	4.70×10^{-5}						
00091-20-3	Naphthalene**	-	-	-	-	-	-	-	-	-	-	-	-						
00095-47-6	o-Xylene	4.47×10^{-4}	0.948	1.02×10^{-3}	4.14×10^{-4}	0.937	1.51×10^{-3}	4.25×10^{-4}	0.963	5.18×10^{-4}	2.56×10^{-4}	0.982	3.85×10^{-9}						
00095-63-6	1,2,3-Trimethylbenzene	1.69×10^{-4}	0.962	5.57×10^{-4}	1.43×10^{-4}	0.923	2.30×10^{-3}	1.57×10^{-4}	0.965	4.58×10^{-4}	7.31×10^{-5}	0.974	2.02×10^{-8}						
00098-82-8	Isopropylbenzene**	1.48×10^{-4}	0.950	9.40×10^{-4}	1.28×10^{-4}	0.920	2.48×10^{-3}	1.21×10^{-4}	0.906	3.39×10^{-3}	-	-	-						
00100-41-4	Ethyl Benzene**	5.79×10^{-4}	0.979	1.64×10^{-4}	5.20×10^{-4}	0.980	1.46×10^{-4}	6.28×10^{-4}	0.992	2.60×10^{-5}	2.86×10^{-4}	0.973	2.29×10^{-8}						
00100-42-5	Styrene**	1.34×10^{-4}	0.979	1.64×10^{-4}	1.16×10^{-4}	0.980	1.46×10^{-4}	1.07×10^{-4}	0.992	2.60×10^{-5}	7.39×10^{-5}	0.982	4.10×10^{-9}						
00103-65-1	n-Propylbenzene	2.68×10^{-4}	0.995	7.98×10^{-6}	-	-	-	2.24×10^{-4}	0.988	5.49×10^{-5}	1.11×10^{-4}	0.915	4.10×10^{-6}						
00106-42-3	p-Xylene	4.56×10^{-4}	0.979	1.64×10^{-4}	4.08×10^{-4}	0.980	1.46×10^{-4}	3.94×10^{-4}	0.968	3.87×10^{-4}	2.34×10^{-4}	0.967	6.02×10^{-8}						
00107-83-5	2-Methylpentane	-	-	-	6.04×10^{-4}	0.980	1.46×10^{-4}	-	-	-	-	-	-						
00108-10-1	MIBK**	3.61×10^{-3}	0.929	1.94×10^{-3}	3.48×10^{-3}	0.980	1.46×10^{-4}	3.01×10^{-3}	0.992	2.60×10^{-5}	3.16×10^{-3}	0.989	4.70×10^{-5}						
00108-38-3	m-Xylene	5.96×10^{-4}	0.979	1.64×10^{-4}	5.35×10^{-4}	0.980	1.46×10^{-4}	5.56×10^{-4}	0.956	7.34×10^{-4}	3.00×10^{-4}	0.971	3.23×10^{-8}						
00108-65-6	PGMEA	3.99×10^{-2}	0.991	4.41×10^{-5}	2.33×10^{-2}	0.961	1.46×10^{-4}	5.77×10^{-1}	0.875	6.00×10^{-3}	3.21×10^{-1}	0.988	5.49×10^{-5}						
00108-67-8	1,3,5-Trimethylbenzene	1.81×10^{-4}	0.995	7.83×10^{-6}	1.38×10^{-4}	0.979	1.66×10^{-4}	1.70×10^{-4}	0.992	2.60×10^{-5}	5.64×10^{-5}	0.978	9.91×10^{-9}						
00108-88-3	Toluene	4.49×10^{-3}	0.661	4.91×10^{-2}	4.22×10^{-3}	0.854	8.40×10^{-3}	2.14×10^{-3}	0.819	1.31×10^{-2}	2.41×10^{-3}	0.716	1.02×10^{-3}						
00110-54-3	n-Hexane	3.75×10^{-3}	0.998	1.08×10^{-6}	3.67×10^{-3}	0.992	2.43×10^{-3}	3.30×10^{-3}	0.992	2.60×10^{-5}	2.36×10^{-3}	0.453	2.32×10^{-2}						
00111-84-2	n-Nonane	-	-	-	-	-	-	-	-	-	7.17×10^{-5}	0.982	1.22×10^{-4}						
00112-40-3	n-Dodecane	9.70×10^{-5}	0.985	8.69×10^{-5}	9.10×10^{-5}	0.980	1.46×10^{-4}	8.53×10^{-5}	0.992	2.60×10^{-5}	2.00×10^{-4}	0.973	2.44×10^{-8}						
00141-78-6	Ethyl Acetate	4.15×10^{-3}	0.979	1.64×10^{-4}	5.84×10^{-3}	0.980	1.46×10^{-4}	-	-	-	3.50×10^{-3}	0.989	4.70×10^{-5}						
00526-73-8	1,2,4-Trimethylbenzene	3.22×10^{-4}	0.994	1.44×10^{-5}	2.50×10^{-4}	0.980	1.47×10^{-4}	2.50×10^{-4}	0.954	8.14×10^{-4}	8.05×10^{-5}	0.982	3.36×10^{-9}						
00611-14-3	o-Ethyltoluene	1.49×10^{-4}	0.979	1.64×10^{-4}	2.26×10^{-4}	0.980	1.46×10^{-4}	2.45×10^{-4}	0.992	2.60×10^{-5}	8.75×10^{-5}	0.968	3.76×10^{-4}						
00620-14-4	m-Ethyltoluene	2.76×10^{-4}	0.992	2.67×10^{-5}	2.22×10^{-4}	0.981	1.30×10^{-4}	2.20×10^{-4}	0.965	4.66×10^{-4}	8.44×10^{-5}	0.987	9.79×10^{-10}						
00622-96-8	p-Ethyltoluene	2.73×10^{-4}	0.992	2.58×10^{-5}	2.36×10^{-4}	0.986	7.33×10^{-5}	2.22×10^{-4}	0.980	1.52×10^{-4}	8.57×10^{-5}	0.990	2.44×10^{-10}						
01120-21-4	n-Undecane	-	-	-	-	-	-	-	-	-	5.45×10^{-5}	0.982	1.22×10^{-4}						
01634-04-4	MTBE	-	-	-	8.35×10^{-3}	0.980	1.46×10^{-4}	-	-	-	4.91×10^{-3}	0.989	4.70×10^{-5}						

PGMEA: Propylene glycol monomethyl ether acetate; MIBK: 4-Methyl-2-Pentanone; MTBE: Methyl-tert-Butylether.

*: IARC group 2A; **: IARC group 2B.

“-”: Non-detected.

Table 5. Year-long concentrations of each individual VOC (C_{voc} ; ppb) (Mean \pm SD) for the eight studied areas using their corresponding predictive models.

CAS No.	Chemical	Array								CF				Cell
		CVD	Dry	Wet	Photo	ITO	BM/R	G/B	PI					
00064-17-5	Ethanol	1199 \pm 668	1241 \pm 743	839 \pm 584	863 \pm 573	520 \pm 181	1324 \pm 726	838 \pm 402	3117 \pm 1315					
00067-63-0	2-Propanol	-	19.6 \pm 11.7	24.3 \pm 16.9	35.5 \pm 23.6	7.89 \pm 2.75	94.3 \pm 51.7	-	37.1 \pm 15.6					
00067-64-1	Acetone	332 \pm 185	255 \pm 152	236 \pm 164	218 \pm 144	425 \pm 148	1095 \pm 601	1075 \pm 516	650 \pm 274					
00075-09-2	Methylene Chloride*	-	32.1 \pm 19.2	54.1 \pm 37.6	83.1 \pm 55.2	31.2 \pm 10.9	33.4 \pm 18.3	58.7 \pm 28.2	12.9 \pm 5.46					
00075-69-4	Trichlorofluoromethane	-	-	-	-	2.63 \pm 0.92	3.49 \pm 1.92	-	-					
00076-13-1	1,1,2-Trichlorotrifluoroethane	-	10.7 \pm 6.39	14.8 \pm 10.3	-	-	-	-	-					
00078-78-4	Isopentane	-	-	-	12.6 \pm 8.37	-	-	-	-					
00078-93-3	Butanone	-	15.4 \pm 9.23	17.5 \pm 12.2	19.9 \pm 13.2	6.87 \pm 2.39	-	-	-				12.7 \pm 5.37	
00091-20-3	Naphthalene**	-	-	-	11.2 \pm 7.44	-	-	-	-					
00095-47-6	o-Xylene	2.04 \pm 1.14	16.4 \pm 9.84	2.09 \pm 1.45	2.23 \pm 1.48	0.72 \pm 0.25	1.03 \pm 0.57	0.98 \pm 0.47	13.2 \pm 5.58					
00095-63-6	1,2,3-Trimethylbenzene	0.53 \pm 0.30	0.51 \pm 0.30	0.52 \pm 0.36	0.53 \pm 0.35	0.27 \pm 0.09	0.36 \pm 0.19	0.36 \pm 0.17	0.27 \pm 0.11					
00098-82-8	Isopropylbenzene**	0.47 \pm 0.26	0.45 \pm 0.27	0.47 \pm 0.33	0.48 \pm 0.32	0.24 \pm 0.08	0.32 \pm 0.18	0.28 \pm 0.13	-					
00100-41-4	Ethyl Benzene**	3.96 \pm 2.21	3.86 \pm 2.31	4.27 \pm 2.97	3.49 \pm 2.32	0.94 \pm 0.33	1.29 \pm 0.71	1.45 \pm 0.69	1.04 \pm 0.44					
00100-42-5	Styrene**	-	0.42 \pm 0.25	0.45 \pm 0.31	0.46 \pm 0.30	0.22 \pm 0.08	0.29 \pm 0.16	0.25 \pm 0.12	0.27 \pm 0.11					
00103-65-1	n-Propylbenzene	0.64 \pm 0.36	1.98 \pm 9.84	0.65 \pm 0.45	0.65 \pm 0.43	0.43 \pm 0.15	-	0.52 \pm 0.25	0.93 \pm 0.39					
00106-42-3	p-Xylene	2.11 \pm 1.17	2.54 \pm 1.52	2.09 \pm 1.45	2.10 \pm 1.39	0.74 \pm 0.26	1.02 \pm 0.56	0.91 \pm 0.43	0.85 \pm 0.36					
00107-83-5	2-Methylpentane	-	-	-	-	-	1.51 \pm 0.83	-	-					
00108-10-1	MIBK**	-	14.2 \pm 8.51	16.2 \pm 11.3	16.8 \pm 11.1	5.83 \pm 2.03	8.68 \pm 4.76	6.93 \pm 3.32	11.5 \pm 4.84					
00108-38-3	m-Xylene	4.16 \pm 1.17	3.86 \pm 2.31	4.49 \pm 3.13	4.70 \pm 3.12	0.96 \pm 0.34	1.33 \pm 0.73	1.28 \pm 0.61	1.09 \pm 0.46					
00108-65-6	PGMEA	32.8 \pm 15.9	102 \pm 72.2	21.6 \pm 14.2	3390 \pm 2250	64.6 \pm 24.8	58.3 \pm 41.9	1328 \pm 637	1165 \pm 979					
00108-67-8	1,3,5-Trimethylbenzene	0.49 \pm 0.27	0.49 \pm 0.29	0.51 \pm 0.36	0.52 \pm 0.35	0.29 \pm 0.10	0.34 \pm 0.19	0.39 \pm 0.19	0.21 \pm 0.09					
00108-88-3	Toluene	12.2 \pm 6.82	3.97 \pm 2.38	21.3 \pm 14.8	8.45 \pm 5.61	7.26 \pm 2.53	10.5 \pm 5.77	4.92 \pm 2.36	1.09 \pm 0.46					
00110-54-3	n-Hexane	-	0.61 \pm 0.37	3.64 \pm 2.53	13.7 \pm 9.09	6.06 \pm 2.11	9.15 \pm 5.02	7.59 \pm 3.64	0.40 \pm 0.17					
00111-84-2	n-Nonane	-	-	0.23 \pm 0.16	-	-	-	-	0.19 \pm 0.08					
00112-40-3	n-Dodecane	0.76 \pm 0.36	2.78 \pm 1.66	0.89 \pm 0.62	0.62 \pm 0.41	0.16 \pm 0.05	0.23 \pm 0.12	0.19 \pm 0.09	0.73 \pm 0.31					
00141-78-6	Ethyl Acetate	-	3.97 \pm 2.38	16.9 \pm 11.8	38.7 \pm 25.7	6.71 \pm 2.34	14.6 \pm 7.98	-	8.76 \pm 3.69					
00526-73-8	1,2,4-Trimethylbenzene	0.99 \pm 0.55	0.89 \pm 0.54	0.91 \pm 0.64	0.01 \pm 0.69	0.52 \pm 0.18	0.62 \pm 0.34	0.58 \pm 0.28	0.29 \pm 0.12					
00611-14-3	o-Ethyltoluene	0.49 \pm 0.27	20.2 \pm 12.1	0.51 \pm 0.35	0.52 \pm 0.34	0.24 \pm 0.08	0.56 \pm 0.31	0.56 \pm 0.27	17.8 \pm 7.53					
00620-14-4	m-Ethyltoluene	0.73 \pm 0.40	0.48 \pm 0.29	0.74 \pm 0.51	0.75 \pm 0.49	0.45 \pm 0.16	0.55 \pm 0.30	0.51 \pm 0.24	0.32 \pm 0.13					
00622-96-8	p-Ethyltoluene	0.68 \pm 0.38	0.69 \pm 0.42	0.65 \pm 0.45	0.67 \pm 0.45	0.44 \pm 0.15	0.59 \pm 0.32	0.51 \pm 0.25	0.31 \pm 0.13					
01120-21-4	n-Undecane	-	-	0.28 \pm 0.19	-	-	-	-	8.57 \pm 3.62					
01634-04-4	MTBE	-	0.64 \pm 0.38	25.3 \pm 17.6	22.7 \pm 15.1	-	20.8 \pm 11.4	-	0.26 \pm 0.11					

Note: PGMEA: Propylene glycol monomethyl ether acetate; MIBK: 4-Methyl-2-Pentanone; MTBE: Methyl-tert-Butylether.

*: IARC group 2A; **: IARC group 2B.

"-": non-predicted.

Table 6. The estimated long-term total exposure based on the predicted year-long C_{VOC_i} for the eight selected areas.

Process	Area	Total Exposure
Array	CVD	1.94×10^{-3}
	Dry	3.45×10^{-3}
	Wet	3.86×10^{-3}
	Photo	5.68×10^{-3}
CF	ITO	2.17×10^{-3}
	BM/R	4.59×10^{-3}
	G/B	3.88×10^{-3}
Cell	PI	5.34×10^{-3}

in chemical compositions among the eight selected areas (Table 1). While using portable PID as a tool for monitoring, it is known that different VOCs with the same concentration might lead to different measured C_{T-VOCs} levels. Therefore, it is not so surprising to see that higher year-long C_{T-VOCs} (Table 3) were found with lower C_{VOC_i} (Table 5) in some selected areas.

Long-Term Multiple Chemical Exposures for the Eight Selected Areas

Table 6 summarizes the long-term multiple chemical exposures for the eight studied areas based on the estimated year-long C_{VOC_i} . For the array process, the long-term multiple chemical exposures were found as 1.94×10^{-3} , 3.45×10^{-3} , 3.86×10^{-3} and 5.68×10^{-3} for the areas of the CVD, Dry, Wet, and Photo, respectively. For ITO, BM/R, and G/B of the CF process, the exposure levels were found as 2.17×10^{-3} , 4.59×10^{-3} , and 3.88×10^{-3} , respectively. For the PI area of the Cell process, the exposure level of 5.34×10^{-3} was obtained. The levels found in the areas of the Photo and PI were respectively 1.2–2.9 and 1.2–2.8 folds in magnitude higher than that of other areas. The above results could be because high amounts of PGMEA and ethanol were used in the Photo and PI area, respectively. The long-term multiple chemical exposures for the eight studied areas fell to the range 0.1%–0.6% of the permissible exposure level (= 1).

Estimating Acceptable C_{T-VOCs} Levels for the Eight Selected Areas

Considering measuring C_{VOC_i} using traditional sorbent tube methods is expensive and labor intensive, and hence it would be difficult for enterprises to measure C_{VOC_i} on a routine basis in order to obtain a large amount long-term exposure data. Since C_{T-VOCs} measured by the portable PID can be used for effectively predicting C_{VOC_i} , particularly the involved measuring method is simple and less expensive, the use of C_{T-VOCs} as a surrogate for the purpose of routinely checking the exposure conditions could be theoretically plausible. Using the regression models obtained from the present study, Table 7 shows the recommended acceptable C_{T-VOCs} levels for each of the eight selected areas, assuming the acceptable total exposure is set at 1.0. The resultant lowest and highest allowable C_{T-VOCs} are found for Array/CVD (49.1 ppm), and Array/photo (577 ppm), respectively. Apparently, the selected area with a higher total exposure level would be accompanied with a lower allowable C_{T-VOCs} .

Table 7. Recommended acceptable C_{T-VOCs} levels (ppm) for each of the eight selected areas, assuming the acceptable total exposure is set at 1.0.

Process	Area	Acceptable C_{T-VOCs}
Array	CVD	49.1
	Dry	300
	Wet	471
	Photo	577
CF	ITO	337
	BM/R	193
	G/B	250
Cell	PI	56.3

Since there are intrinsic differences in chemical compositions among the eight selected areas (Table 1), and it is known that different VOCs with the same concentration might lead to different measured C_{T-VOCs} levels, it is not so surprising to see that the above inference contradicts the results found in Table 6 (i.e., the total exposure level) and Table 7 (i.e., the allowable C_{T-VOCs}).

CONCLUSIONS

The present study found that the concentrations of C_{VOC_i} for the eight selected areas were much lower than their corresponding PEL-TWAs. High C_{VOC_i} were found in those areas using ethanol, acetone, and PGMEA as their raw materials. By relating C_{VOC_i} to C_{T-VOCs} , results show that all resultant regression coefficients were statistically significant, and the all regression models were found with high R^2 indicating that C_{T-VOCs} was adequate for predicting C_{VOC_i} . The estimated long-term total exposures for all studied areas fell to the range 0.10%–20% of the acceptable levels. Using the C_{T-VOCs} as a surrogate for the routine checking purpose, the present study yielded allowable C_{T-VOCs} falls to the ranges of 49.1 ppm–577 ppm. Considering the approach used in the present study requires less cost and manpower, and hence it can be regarded as a useful tool for conducting long-term multiple chemical exposure assessments for other similar industries in the future.

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