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Impacts of COVID-19's restriction measures on personal exposure to VOCs and aldehydes in Taipei City

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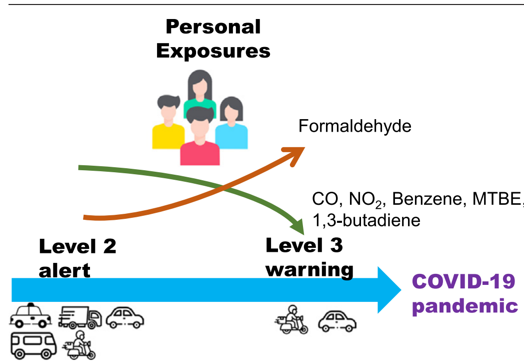
HIGHLIGHTS

- Benzene and 1,3-butadiene exposure decreased during stricter control measures.
- Exposure to formaldehyde was increased due to stricter restriction measures.
- The abatement of transportation can reduce personal exposure to specific VOCs.
- A significant reduction (42 %) in cancer risks for VOC exposure was observed.
- Traffic flow reductions confirm the decrease in traffic-related VOCs.

GRAPHICAL ABSTRACT



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ABSTRACT

The Coronavirus Disease 2019 (COVID-19) pandemic provided an unprecedented natural experiment, that allowed us to investigate the impacts of different restrictive measures on personal exposure to specific volatile organic compounds (VOCs) and aldehydes and resulting health risks in the city. Ambient concentrations of the criteria air pollutants were also evaluated. Passive sampling for VOCs and aldehydes was conducted for graduate students and ambient air in Taipei, Taiwan, during the Level 3 warning (strict control measures) and Level 2 alert (loosened control measures) of the COVID-19 pandemic in 2021–2022. Information on the daily activities of participants and on-road vehicle counts nearby the stationary sampling site during the sampling campaigns were recorded. Generalized estimating equations (GEE) with adjusted meteorological and seasonal variables were used to estimate the effects of control measures on average personal exposures to the selected air pollutants. Our results showed that ambient CO and NO_2 concentrations in relation to on-road transportation emissions were significantly reduced, which led to an increase in ambient O_3 concentrations. Exposure to specific VOCs (benzene, methyl tert-butyl ether (MTBE), xylene, ethylbenzene, and 1,3-butadiene) associated with automobile emissions were remarkably decreased by ~40–80 % during the Level 3 warning, resulting in 42 % and 50 % reductions of total incremental lifetime cancer risk (ILCR) and hazard index (HI), respectively, compared with the Level 2 alert. In contrast, the exposure concentration and calculated health risks in the selected population for formaldehyde increased by ~25 % on average during the Level 3 warning. Our study improves knowledge of the influence of a series of anti-COVID-19 measures on personal exposure to specific VOCs and aldehydes and its mitigations.

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1. Introduction

Volatile organic compounds (VOCs, including aldehydes) are ubiquitous in the environment and not only cause adverse health outcomes through the inhalation route but are also involved in photochemical reactions with oxidants in the atmosphere to produce ozone (O_3) and secondary organic aerosols (SOAs) (Delfino et al., 2003; Sato et al., 2010; D'Andrea and Reddy, 2018; McDonald et al., 2018). For instance, exposure to traffic-related VOCs is associated with acute changes in lung function, inflammation, and heart rate variability among urban cyclists (Weichenthal et al., 2012); and specific VOCs (including alkenes, alkynes and ketone groups) were found to be associated with emergency department visits for cardiovascular diseases and asthma among patients living in Atlanta in the United States (Ye et al., 2017). Exposure to VOCs in cities has become a severe concern owing to their high population densities and traffic volumes. Urban VOCs are typically a complex mix of emissions from various anthropogenic sources, including traffic and industrial emissions, solvent uses, waste burning, and residential activities. In addition, fugitive VOC emissions from restaurants and temples in the Chinese community have been recognized as significant emission sources (Wang et al., 2007; Wang et al., 2018). Strategies to improve air quality and mitigate human exposure to urban VOCs are needed to minimize potential health impacts. Although most anthropogenic sources of urban VOCs have been recognized, it is difficult to demonstrate the effectiveness of specific control measures (i.e., reductions in transportation proximity to industrial and commercial sectors) in reducing personal exposure to urban VOCs.

Since the first case of novel Coronavirus Disease 2019 (COVID-19) was reported in Wuhan, China, in December 2019, the COVID-19 epidemic quickly spread across the world resulting in a global pandemic. To mitigate this outbreak, many countries have implemented social and economic restrictions, such as working from home, distanced education, travel ban, and closed entertainment venues. Many countries locked down city residents and enforced strict quarantine to control the spread of this highly communicable disease. As a result, people have changed their lifestyles and daily activities and spent more time at home. While most people in the world are at risk of exposure to severe acute respiratory syndrome coronavirus 2 (SARS-CoV-2), a positive impact of the pandemic on ambient air quality has been reported in many countries, leading to significant abatement in the concentrations of outdoor particulate matter, NO_2, CO, SO_2, and VOCs (Adam et al., 2021; Amouei Torkmahalleh et al., 2021; Le et al., 2020; Venter et al., 2020). Thus, the COVID-19 pandemic provides an unprecedented natural and controlled experiment that can improve our understanding of changes in air quality and its causes.

The reduction in ambient VOC concentrations based on stationary measurements has been evaluated during the outbreak control period (Jia et al., 2020; Guevara et al., 2021; Wang et al., 2021; Pei et al., 2022; Zhang et al., 2022). Ambient concentrations of aromatics (i.e., benzene, toluene, ethylbenzene, and xylene) and alkenes with a significant decline (up to 80 %) in the urban areas during the lockdown were also reported, as compared with the pre-lockdown period. However, no studies have assessed personal exposure to specific VOCs and aldehydes, such as benzene and formaldehyde, among city residents, which can directly reflect the impact of air pollutant abatement owing to a series of outbreak controls in cities.

In this study, we evaluated the impacts of restriction measures due to COVID-19 pandemic on ambient air pollutants and personal exposure to VOCs and aldehydes in Taipei City, Taiwan, using prospective exposure assessment. This builds on previous studies that have investigated air pollutants in indoor, outdoor, or ambient environments during the lockdown period using retrospective methods (compared with the historical periods). We developed generalized estimating equations (GEEs) to examine the effects of meteorological variables on personal exposure, and determined the exposure concentrations of specific VOCs and aldehydes for the study population. Overall, our study provides a comprehensive assessment of the health risks associated with inhalation exposure to multiple hazardous air pollutants in the urban area.

2. Materials and methods

2.1. Study setting

Ambient concentrations and personal exposures to VOCs and aldehydes were investigated in Taipei City, Taiwan, during the Level 3 warning and Level 2 alert of the COVID-19 pandemic in 2021and 2022. Fig. 1 shows the timeline of the prevention measures for the COVID-19 epidemic announced by the Central Epidemic Command Center of Taiwan (CECC) in parallel with our campaign. Since the first case of the COVID-19 was reported in Taiwan on Jan. 20, 2020, a series of control measures to prevent a wider outbreak was issued, such as border controls and public transportation restrictions. Owing to severe local infections, the CECC subsequently announced the nationwide Level 3 epidemic alert on May 15, 2021. Disease prevention and control implemented under the Level 3 warning were as follows: (1) members of the public were asked to wear masks all the time when going out; (2) leisure and entertainment venues were ordered to close; (3) food and beverage vendors only offered takeout services; (4) people were urged to reduce the frequency of visiting supermarkets and stores; (5) wedding banquets and public memorial ceremonies of funerals were banned; (6) religious gatherings were completely suspended and religious places were temporarily closed to the public; (7) leisure, exhibition, and entertainment venues were closed, and sports competitions were banned; and (8) educational facilities were closed, with online learning mandatory for students below the high school level. According to individual company policies, some office workers were also required to work from home. Great Taipei City was the area that was most affected by the COVID-19 outbreak.

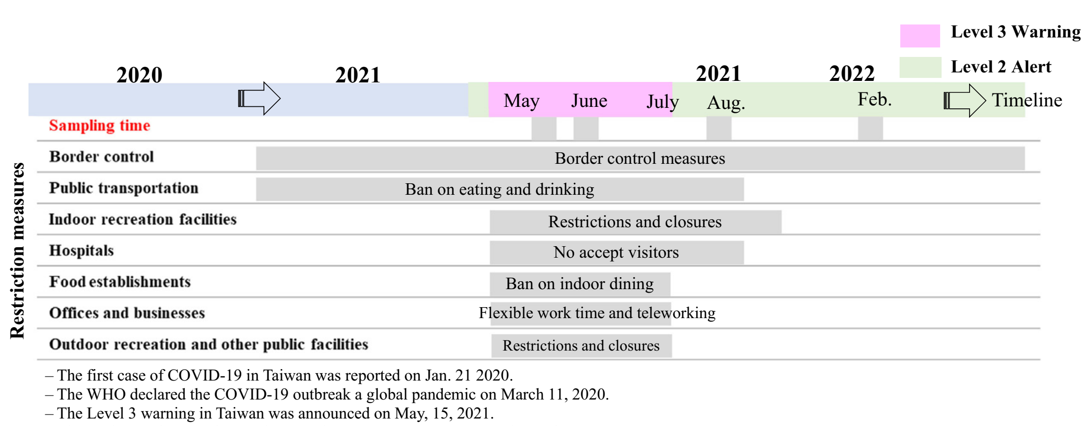

Fig. 1. Timeline of the restriction measures for COVID-19 pandemic in Taiwan.

The CECC lowered the epidemic alert to Level 2 on July 27, 2021, as the epidemic gradually slowed down. The control measures were appropriately adjusted and relaxed as follows: (1) leisure and entertainment venues, stores, and supermarkets were reopened; (2) people were asked to strictly follow rules for measuring body temperature and wearing masks; (3) the catering industry provided in-house food service; (4) religious places were reopened; (5) wedding banquets and public memorial services were allowed again; and (6) educational facilities and venues were reopened, but crowd-control and capacity limit management were required at commercial and public venues. Level 3 warnings during the COVID-19 pandemic had stricter prevention measures than Level 2 alerts. There were no Level 4 warning issued and associated measures (such as lockdown, prohibition of public events, closure of schools, universities, and non-essential business; and closures of external borders with restricted travel and public transport) during the pandemic in Taiwan from 2019 to 2022.

2.2. Study subjects

To assess personal exposure to VOCs and aldehydes affected by these anti-COVID-19 measures, we recruited graduate students (n = 36) from two universities (National Taiwan University and National Yang-Ming Chiao-Tung University) in Taipei, Taiwan. National Taiwan University is located in the Daan District of Taipei, adjacent to an 8-lane road. National Yang-Ming Chiao-Tung University is located in the Beitou District of Taipei. Participants wore personal samplers to collect VOCs and aldehydes for three days during the Level 3 warning (May 27–Jun. 3 and Jun. 28–Jul. 5, 2021) and Level 2 alert (Aug. 10–16, 2021, and Feb. 22–Mar. 1, 2022) of the COVID-19 pandemic. The participants were also asked to record their activity patterns, such as time spent indoors, cooking, smelling odors, and use of cosmetics. All participants are non-smokers.

2.3. VOCs and aldehydes sampling

Two passive diffusive samplers (VOCs and aldehydes) were worn simultaneously in each participant's breathing zone for the three-day study period. Two repeated measurements were performed for each participant across weekdays (Monday to Wednesday) and weekends (Friday to Sunday) for each campaign (Level 2 vs. Level 3). No restrictions on personal behavior were prescribed during the sampling period. Passive samplers (VOCs and aldehydes) were also placed at a nearby fixed station in the National Taiwan University to collect ambient concentration data during personal sampling. A VOC sampler (ULTRA Passive Samplers, SKC Inc., USA) with 500 g charcoal and an aldehyde sampler with cartridge adsorbents (RAD165, Radiello, Merck Inc., Germany) with 2,4-DNPH coated FLORISIL were used. After sampling, both types of samplers were tightly sealed and stored at 4 °C when shipped to the laboratory. All samples were then stored at −20 °C prior to the subsequent analysis. Field blanks were also conducted in each campaign and followed the same protocol.

2.4. VOC and aldehydes analysis

The VOC samples were analyzed at the certified laboratory of the National Health Research Institutes. After defrosting the samples to a normal temperature, we poured the activated carbon with any absorbed VOCs into an extraction bottle and added 100 μL of internal standards with a concentration of 0.5 μg/mL. Next, 2 mL of dichloromethane was added, and the mixture was shaken for 30 min. The extracts were filtered through a 13-mm PTFE membrane (0.2 μm pore size, Acrodisc® syringe filters, Merck Inc., Germany) and stored in gray bottles for analysis. The extract samples were analyzed using a gas chromatographer (GC, Agilent Technologies 7890B) coupled with mass spectrometric detection (MS-MS triple Guard, Agilent Technologies 7000C) in multiple reaction monitoring (MRM) mode. The injection port used an Agilent liner 5190-2295 split tube (split 5:1, split slow 7 mL/min) at a temperature of 120 °C. Helium was used as the carrier gas, the front-end flow was 1.4 mL/min, and the back-end flow was 2.4 mL/min. The oven temperature program was held for 2 min at 32 °C, increased to 110 °C at a rate of 20 °C/min, and then heated to 230 °C at a rate of 40 °C/min and hold 2 min. Finally, the oven was programed maintain a constant temperature of 230 °C to clean the column using helium back flushing for 2 min.

The capillary column (DB-624 UI, length = 30 m, inner diameter = 0.32 mm, film thickness = 1.8 μm) and an uncoated capillary column (length = 1.19 m, inner diameter = 0.15 mm, film thickness = 0 μm) were used for compounds separation. The temperature of the hexapole column (collision cell) and ion source was 250 °C, and the temperature of MS 1 and MS 2 was 150 °C. Thirteen VOC standards (acrylonitrile, 1,3-butadiene, methyl tert-butyl ether, (MTBE), ethyl acetate, Chloroform, 1,2-dichloroethane, benzene, toluene, chlorobenzene, ethylbenzene, m/p-xylene, o-xylene, and 1,2-dichlorobenzen) plus internal standards (acrylonitrile-d3, methyl tert-butyl ether-d3, 1,2-dichloroethane-d_4, benzene-d_6, toluene-d_8, chlorobenzene-d_5, ethyl benzene-d_{10}, o-xylene-d_6, and 1,2-dichlorobenzene-d_4) were analyzed within a concentration range of 0.5–100 ng/mL to establish standard calibration curves. Table S1 provides detailed results of quality assurance and quality control (QA/QC); the low limit of detection (LOD) ranged from 0.064 to 1.64 ng for the selected compounds. Known amounts of the VOC mixture standard mixtures were added to blank medians and analyzed using the same procedures to evaluate the recovery rate. The recovery efficiencies of all compounds ranged from 85.6 % to 110 %.

Aldehydes derivatives were extracted with acetonitrile (2 mL) for 30 min and analyzed using high-performance liquid chromatography (HPLC, PU-2089, Jasco, Japan) combined with an ultraviolet detector (UV, Varian ProStar 320, Varian, USA) at a detection wavelength of 360 nm. Chromatographic separation was carried out in a reverse-phase column (Ascentis® RP-Amide Column, 5 μm, 250 × 4.6 mm, Supelco, Bellefonte, Pennsylvania, USA) with a gradient mobile phase of acetonitrile/water. The gradient program was performed at a 1.2 mL/min flow rate as follows: an initial acetonitrile/water ratio of 40/60 for 1 min, an increase in acetonitrile from 40 % to 65 % within 24 min, held for 5 min, an increase in acetonitrile from 65 % to 95 % within 28 min, held for 3 min, a decrease in acetonitrile from 95 % to 40 % within 10 min, and then held for 3 min (73 min in total). Nine aldehydes (formaldehyde, acetaldehyde, acrolein, propionaldehyde, butyraldehyde, benzaldehyde, isovaleraldehyde, valeraldehyde, and hexaldehyde) were analyzed. Table S1 provides the results of the QA/QC, which were quantitatively characterized by the acceptable recovery rate (76.4 %–111 %) and reproducibility (relative standard deviation (RSD) < 2.5 %). Field blanks were extracted and analyzed every 10 samples. All samples were subtracted from the background value of the blank adsorbents. When the value of VOCs and aldehydes for a given sample fell below the LOD value the corresponding sample was assigned a value of LOD/2 for that compound. Table S2 shows the percentage of samples below the LOD for all measurements. Personal exposure and ambient concentrations of the selected VOCs and aldehydes recorded via the passive diffusive samplers were calculated using the following equation.

$$C = \frac{(SW - BW) \times 10^3}{SR \times T} \tag{1}$$

where C is the concentration of air contaminants (μg/m^3); SW is the sample weight (ng) obtained by analysis; BW is the analyte weight in the blank (ng); SR is the sampling rate (mL/min, shown in Table S3); and T is the sampling time (min).

2.5. Air pollutants and meteorological variables

Air pollutant concentrations and meteorological conditions were obtained from the Guting Air Quality Monitoring Station (established by the Taiwan EPA). This monitoring station is 1.72 km from the main campus of National Taiwan University. The observed ambient air pollutants include methane (CH_4), non-methane hydrocarbon (NMHC), total hydrocarbon (THC), carbon monoxide (CO), nitric oxide (NO), nitrogen dioxide (NO_2), nitrogen oxide (NO_X), ozone (O_3), sulfur dioxide (SO_2), particulate matter

(PM) with an aerodynamic diameter ≤ 10 μm (PM_{10}), and PM with an aerodynamic diameter ≤ 2.5 μm ($PM_{2.5}$). Meteorological factors included temperature, rainfall, and relative humidity (RH). The traffic flow, including vehicle classification and counting during the study periods, was also calculated using a computer vision-based vehicle detection and tracking system. The camera device was installed at the interception of Keelung Road and Xinhai Road on the main campus of National Taiwan University. Traffic data extraction from the collected videos consisting of vehicle detection, trajectory tracking, and vehicle counting was implemented using the Python programming language and OpenCV library.

2.6. Health risk assessment

To evaluate the potential health risks resulting from inhalation exposure to VOCs and aldehydes, we calculated the increased lifetime cancer risk (ILCR) for the Level 2 alert and Level 3 warning. The ILCR is calculated by multiplying the measured concentration of a selected compound by its corresponding inhalation unit risk (IUR), as follows:

$$ILCR_i = C_i \times IUR_i \tag{2}$$

where $ILCR_i$ is the increased lifetime *cancer risk associated with compound i*; C_i is the personal exposure concentration (μg/m^3) of the target VOCs or aldehydes; and IUR_i (μg/m^3)$^{-1}$ represents the unit risk for compound i obtained from the Integrated Risk Information System (IRIS), USEPA (https://www.epa.gov/iris). The hazard quotient (HQ) was estimated for potential non-cancer health risks based on the chronic reference exposure limit (REL), as follows:

$$HQ_i = \frac{C_i}{REL_i} \tag{3}$$

where HQ_i is the hazard quotient for compound i, and REL_i is the reference concentration of compound i obtained from the Office of Environmental Health Hazard Assessment (OEHHA), California EPA (https://oehha.ca.gov). $HQ_i \le 1$ indicates no risk to human health, and $HQ_i > 1$ indicates that compound i poses some level of risk to human health. The hazard index (HI) was used to summarize the non-cancer risks from all of the selected compounds, as follows:

$$HI = \Sigma HQ_i \tag{4}$$

2.7. Statistical analysis

Descriptive statistics were used to describe the data for the selected VOCs and aldehydes. Significance testings for comparisons between the Level 2 alert and Level 3 warning for air quality (i.e., NO_2, $PM_{2.5}$, etc.), meteorological conditions, traffic flow, mass rapid transit (MRT) volume, and personal exposures (VOCs and aldehydes) between the Level 2 alert and Level 3 warning was conducted using the paired t-test. The difference in ambient concentrations of selected VOCs and aldehydes between two periods was evaluated using the Mann-Whitney U test. Spearman rank correlation coefficients to examine the strength of correlations between selected VOCs and aldehydes for the Level 2 alert and Level 3 warning were calculated. The USEPA positive matrix factorization (PMF) 5.0 model was utilized to determine the origin of selected VOCs and aldehydes for each period. The PMF approach has been explored in previous studies (Hopke et al., 2020). We also used a GEE model with adjusted meteorological and seasonal variables and recorded activities to evaluate the effects on VOCs and aldehydes (log-transformed concentrations, μg/m^3) for the Level 3 warning compared with the Level 2 alert. The GEE model was as follows:

$$Y = \beta_0 + \beta_1 X_1 + \ldots + \beta_n X_n + \varepsilon \tag{5}$$

where, Y is the log-transformed VOC or aldehyde concentration (μg/m^3); β_0 is the overall intercept for the group corresponding to the mean background level (log-transformed) when all factors are equal to zero; X_1 to X_n represent the independent variables (such as restrictive levels, temperature, RH, season, and activities); B_1 to B_n are fixed coefficients; and ε is a random error. All statistical analyses were performed using IBM SPSS 22 software (SPSS Inc., IBM Company, Chicago, IL, USA).

3. Results

3.1. Ambient criteria air pollutants

Descriptive statistics of daily air pollutant concentrations and meteorological parameters for the Level 2 alert and Level 3 warning during the study period are shown in Table S4. To limit the effects of temperature and seasonal variations, the criteria air pollutants were obtained at the fixed monitoring station under the similar meteorological condition. We found that the mean (here referred to as arithmetic mean, AM) concentrations of gaseous pollutants (such as CH_4, THC, CO, NO, NO_2, and NO_X) were significantly lower for the Level 3 warning than for the Level 2 alert. There was no significant difference between the two Levels for $PM_{2.5}$ and PM_{10}. In contrast, significantly high concentrations of SO_2 and NMHC were observed for the Level 3 warning. Although the mean concentration of O_3 for the Level 3 warning (26.0 ppb) was higher than for the Level 2 alert (24.0 ppb), this was not statistically significant.

3.2. VOC and aldehyde exposures

Table 1 shows the information on subjects' characteristics and the percentage of their time spent (mean ± SD) on selected daily activities during the sampling periods. The number of subjects, and subjects' age and gender between the Level 2 alert and Level 3 were similar. We found that the subjects had a higher percentage of time spent staying at an indoor home and cooking at home during Level 3 warning and using cosmetics during Level 2 alert. Table 2 shows the descriptive statistics for exposure to VOCs and aldehydes among the graduate students for the Level 2 alert and Level 3 warning sampling periods. The ambient concentrations of VOCs collected during the same periods of personal monitoring are also presented in Table S5. Most of the selected VOCs and aldehydes were detectable in personal and ambient concentrations, with values greater than the LOD. Only acrylonitrile, 1,2-dichlorobenzene, 1,2-dichloroethane, and isovaleraldehyde, which are related to industrial processes and petrochemical emissions, had 30 % measurements with concentrations below the LOD, and, therefore, were not considered further. We found that the subjects had a lower exposure to total VOCs (AM = 36.3 μg/m^3) for the Level 3 warning than the Level 2 alert (AM = 41.7 μg/m^3), although there was no significant difference (p-value = 0.345) as shown in Table 2. We also observed a similar trend for BTEX (the sum of benzene, toluene, ethylbenzene, and xylene) with insignificant difference between two restrictive levels (32.1 μg/m^3 for Level 2 vs. 27.7 μg/m^3 for Level 3, p-value = 0.457). Toluene was the predominant compound, accounting for ~50 % of the total VOCs in the

Table 1
The information on subjects' characteristics and the percentage of their time spent (mean ± SD) on selected daily activities during the sampling periods.

Valuables	Level 2 alert	Level 3 warning	p-Value
Characteristics			
Involved schools	NTU and NYMCTU	NTU and NYMCTU	–
Number of subjects	26	24	–
Age	22–24	22–24	–
Gender (female), %	48.7	55	0.519[b]
Activities			
Stay indoor time, %	59.6 ± 23.3	68.5 ± 24.8	0.017[a]
Cooking at home (yes), %	47.3 ± 42.6	67.5 ± 43.6	0.014[b]
Smell odors (yes), %	63.5 ± 46.3	63.8 ± 41.7	>0.999[b]
Cosmetics use (yes), %	35.1 ± 36.8	16.3 ± 30.1	0.009[b]

NTU: National Taiwan University, NYMCTU: National Yang-Ming Chiao-Tung University.

[a] Mann-Whitney U test.

[b] Fisher's exact test.

Table 2
Descriptive statistics of exposure to VOCs and aldehydes among 34 graduate students for sampling periods of the Level 2 alert and Level 3 warning.

Compounds (μg/m^3)	Level 2 alert (n = 74)							Level 3 warning (n = 80)							*p*-Value*
	AM	SD	GM	IQR	P5	P95	%	AM	SD	GM	IQR	P5	P95	%	
VOCs															
Acrylonitrile	0.050	0.056	0.021	0.072	0.004	0.193	0.12	0.057	0.057	0.028	0.084	0.004	0.184	0.15	0.830
Benzene	2.47	1.90	2.12	1.30	0.966	4.75	5.82	1.33	0.673	1.16	1.01	0.430	2.62	3.58	<0.001
Chlorobenzene	0.018	0.016	0.010	0.022	0.001	0.056	0.04	0.012	0.010	0.008	0.013	0.001	0.031	0.03	0.061
Chloroform	1.44	4.40	0.377	0.956	0.011	4.89	3.39	0.919	1.13	0.714	0.451	0.309	2.43	2.47	0.256
Ethyl acetate	3.54	3.22	2.73	2.60	0.790	8.25	8.35	4.36	3.64	3.45	3.67	1.11	10.9	11.7	0.116
Ethylbenzene	2.02	1.78	1.67	1.08	0.615	3.89	4.76	1.37	0.603	1.23	0.831	0.560	2.54	3.68	0.010
MTBE	3.83	2.79	3.00	2.65	0.869	10.0	9.03	2.97	3.81	2.06	2.03	0.495	8.48	7.98	0.068
m,p-Xylene	5.06	6.63	3.42	4.03	0.869	11.9	11.9	4.25	1.86	3.83	2.81	1.80	7.73	11.4	0.329
o-Xylene	2.67	2.49	2.15	1.63	0.690	4.98	6.29	1.84	0.815	1.65	1.00	0.784	3.63	4.95	0.025
Toluene	20.3	22.5	15.3	10.1	5.08	67.7	47.9	19.6	21.8	14.9	12.4	5.46	52.1	52.7	0.612
1,2-Dichlorobenzene	0.023	0.032	0.005	0.044	0.001	0.092	0.05	0.021	0.043	0.006	0.019	0.001	0.092	0.06	0.539
1,2-Dichloroethane	0.136	0.288	0.057	0.126	0.016	0.442	0.32	0.159	0.162	0.095	0.149	0.016	0.538	0.43	0.363
1,3-Butadiene	0.860	1.81	0.333	0.816	0.019	2.65	2.03	0.309	0.424	0.130	0.413	0.018	1.28	0.83	<0.001
Sum of VOCs	41.7	29.5	34.7	26.9	13.1	111	100	36.3	24.6	31.2	21.4	11.8	72.1	100	0.345
Aldehydes															
Formaldehyde	16.4	13.2	12.9	14.5	4.19	38.9	36.9	21.8	9.41	19.7	14.0	8.69	40.1	40.6	0.016
Acetaldehyde	9.70	6.66	7.89	6.86	2.33	22.6	21.8	11.6	7.15	9.27	7.26	2.89	27.6	21.6	0.019
Acrolein	0.592	0.359	0.525	0.279	0.253	1.21	1.33	0.770	0.602	0.627	0.458	0.251	1.86	1.43	0.046
Propionaldehyde	4.74	5.36	2.97	5.84	0.726	12.0	10.7	8.19	5.44	6.38	5.91	1.30	20.8	15.3	0.006
Butyraldehyde	5.13	4.96	3.89	3.37	1.65	18.6	11.6	3.69	2.96	2.94	1.73	1.17	10.7	6.87	0.158
Benzaldehyde	0.690	0.609	0.529	0.401	0.102	1.80	1.55	0.489	0.409	0.355	0.456	0.044	1.20	0.911	0.240
Isovaleraldehyde	0.038	0.028	0.033	0.000	0.026	0.106	0.086	0.026	0.003	0.026	0.001	0.021	0.028	0.048	0.096
Valeraldehyde	0.964	1.01	0.798	0.478	0.427	1.90	2.17	0.684	0.568	0.579	0.285	0.252	1.55	1.27	0.072
Hexaldehyde	6.175	6.63	4.93	3.90	1.81	12.4	13.9	6.44	4.75	5.54	2.97	2.72	14.3	12.0	0.993
Sum of aldehydes	41.6	29.1	33.2	34.6	8.54	107	100	51.4	21.8	45.6	30.4	10.7	91.8	100	0.044

n: number of measurements, AM: arithmetic mean, SD: standard deviation, GM: geometric mean, IQR: interquartile range, P5: 5th percentile, P95: 95th percentile.

* Paired *t*-test.

exposure concentrations, and was not significantly different between the two restrictive levels. The mean concentrations of exposure to benzene, ethylbenzene, o-xylene, and 1,3-butadiene were significantly lower (p-value < 0.05) for the Level 3 warning than the Level 2 alert. However, exposure to total aldehydes for the Level 3 warning (AM = 51.4 μg/m^3) were significantly higher (p-value = 0.044) than the Level 2 alert (AM = 41.6 μg/m^3). Formaldehyde, acetaldehyde, acrolein, and propionaldehyde accounted for ~80 % of total selected aldehydes in personal exposure concentrations. In particular, Formaldehyde was the predominant compound, accounting for ~40 % of the selected aldehydes, and had a significantly higher concentration for Level 3 warning (AM = 21.8 μg/m^3) than Level 2 alert (AM = 16.4 μg/m^3). A similar trend was also observed in average concentrations of acetaldehyde, acrolein and propionaldehyde.

3.3. Factors affecting exposure

Table S6 shows the spearman correlation coefficients between VOCs and aldehydes for the Level 3 warning and Level 2 alert. A positively high correlation ($r > 0.70$, $p < 0.05$) among benzene, ethylbenzene, MTBE, m/p-xylene, o-xylene, toluene, and 1,3-butadiene for both periods was presented; formaldehyde also positively correlated with propionaldehyde and benzaldehyde for both periods, indicating that those compounds are derived from similar emission sources or contributors. Figs. S1 and S2 show the factor profiles of selected VOC and aldehyde in Taipei during the Level 2 alert and Level 3 warning, respectively. The vehicle exhaust of 33.7 % and 27.8 %, indoor activity and solvent use of 21.4 % and 34.7 %, and mixture and secondary reactions of 44.8 % and 37.4 % for the Level 2 alert and Level 3 warning were identified, respectively.

Fig. 2 shows the effect of the Level 3 warning (Ref.: Level 2 alert) on personal exposure to VOCs and aldehydes using the GEE model. Here, we included several variables of RH, season, week, and recorded activities (cooking, smell orders, cosmetics use, and indoor time) for adjustment in the model to reflect the effects of restrictive levels on personal exposure. We found that personal exposure to benzene, ethylbenzene, m, p-xylene, o-xylene, 1,3-butadiene, butyraldehyde, benzaldehyde, and valeraldehyde among the subjects was significantly reduced during the Level 3 warning, by 16.2 %, 43.0 %, 43.1 %, 47.1 %, 82.6 %, 48.2 %, 39.2, and 62.5 % respectively, while mean exposure concentrations of chloroform and ethyl acetate were increased by 46.3 % and 76.9 %, respectively. Although exposure to formaldehyde and propionaldehyde at average concentrations increased, there was no significant difference between restrictive levels using the GEE model with adjusted variables.

In addition to control measures, relative humidity, temporal variables (weekend and week), and personal activities (cooking, smell the orders, cosmetic using, time of staying indoor) could affect personal exposure to VOCs and aldehydes (Tables S7 and S8). For instance, personal exposure to chlorobenzene, chloroform, ethyl acetate, ethylbenzene, m, p-xylene, o-xylene, 1,3-butadiene, formaldehyde, acetaldehyde, propionaldehyde, butyraldehyde, benzaldehyde, and valeraldehyde with a lower concentration can be observed in the cold season. Smelling odors were associated with increased exposure to benzene, MTBE, toluene, 1,3-butadiene, and acetaldehyde. Cooking may positively or negatively associate with some VOCs and aldehydes. The use of cosmetics may increase exposure to ethyl acetate, o-Xylene, formaldehyde, and valeraldehyde.

3.4. Assessment of health risks

Table 2 shows the total ILCR and HI results based on inhalation exposure to VOCs and aldehydes during Level 2 alert and Level 3 warning the periods. The total ILCR values were obtained from the sum of the cancer risks calculated for the individual VOC (acrylonitrile, benzene, chloroform, 1,2-dichloroethane, and 1,3-butadiene), and aldehyde (formaldehyde and acetaldehyde), as shown in Table S9. The mean total ILCR of VOCs was 7.12×10^{-5} (95th = 3.95×10^{-4}) and 4.14×10^{-5} (95th = 7.54×10^{-5}) for the Level 2 alert and Level 3 warning, respectively. A significant reduction (42 %) in ILCR for VOC exposure was observed. However, the mean total ILCR for aldehyde exposure was significantly higher during the Level 3 warning (3.08×10^{-4}) than during the Level 2 alert (2.34×10^{-4}), presenting a 32 % increase. For chronic non-cancer risks, compared with the Level 2 alert, we found that the Level 3 warning significantly reduced the HI of VOC exposures (by 50 %) but significantly increase the aldehyde exposures (by 31 %).

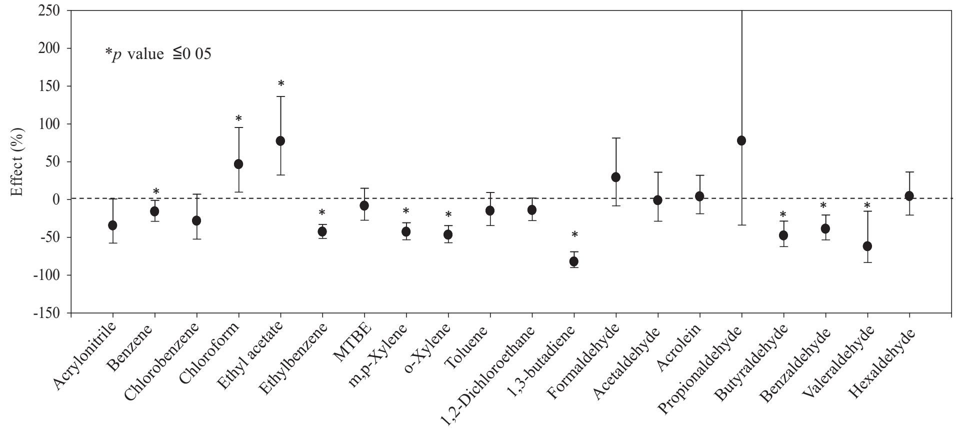

Fig. 2. The effect of the Level 3 warning (Ref.: Level 2 alert) on personal exposure to VOCs and aldehydes (Effect % = (exp(β) − 1) × 100)).

4. Discussion

The decline of air pollutants (such as CO and NO_2) during the Level 3 warning was likely related to stricter restrictions on transportation and social activities. As shown in Table S10, the number of total on-road vehicle counts and MRT passengers during the Level 3 warning significantly decreased by 17.9 % and 65.1 %, respectively, compared with the Level 2 alert. The reductions in vehicle counts per hour for cars, motorbikes, buses, and trucks flows were: 27.9 %, 7.38 %, 28.9 %, and 24.0 %, respectively. Previous studies have reported a reduction in criteria air pollutants' (such as $PM_{2.5}$, PM_{10}, CO, NO_2, SO_2, and O_3) concentrations recorded at fixed monitoring stations or satellite-based observations during lockdown periods in many cities and countries, compared with pre-lockdown periods (Adam et al., 2021; Benchrif et al., 2021; Chen et al., 2020; Chu et al., 2021; Huang et al., 2020; Kanniah et al., 2020; Siciliano et al., 2020). Venter et al. (2020) showed that COVID-19 lockdowns caused a consistent global NO_2 decline (across 34 countries; >10,000 air quality stations) when accounting for meteorological variability. In principle, the significant restriction of urban mobility can lead to a reduction in transport-related pollutants (such as $PM_{2.5}$, NO_2, and CO). Tsai et al. (2021) reported that >90 % of atmospheric CO, 80 % of NOx, and 50 % of $PM_{2.5}$ in Taipei City were attributed to vehicle emissions. Although improvements in urban air quality have been observed in many cities during the pandemic, some studies have reported worsening outdoor air quality among countries during COVID-19 lockdowns. These inconsistent results can be attribute to variations in meteorological factors or the role of atmospheric chemistry and deposition processes (Adam et al., 2021; Pei et al., 2020; Venter et al., 2020). For instance, lockdown measures with a reduction in road traffic and economic activity led to a decrease in $PM_{2.5}$, CO, and NO_2 levels, but an increase in ground-level O_3 concentrations (Amouei Torkmahalleh et al., 2021; Dantas et al., 2020; Li and Tartarini, 2020). Indeed, a significant decrease in NOx emissions from on-road transportations in the cities can lead to an increase in the availability of O_3 (Monks et al., 2015). This phenomenon can also be observed in our study (Table S4). Our study presenting a slight reduction in PMs and an increase in O3 was comparable to the previous studies evaluated during lockdown periods (Bekbulat et al., 2021; Mohd Nadzir et al., 2020; Sicard et al., 2020). In addition, our results showed a relatively high concentration of SO_2 during the Level 3 warning period (5/19–7/26), compared with Level 2 alert (7/27–10/3). SO_2 as a precursor to sulfate plays an important role in the formation of ambient aerosols (Li et al., 2020; Wang et al., 2021), which may be associated with an insignificant difference in PM concentrations between the Level 2 alter and Level 3 warning periods in this study. When we compared SO_2 concentrations during the same periods (5/19–7/26 vs. 7/27–10/3) in 2018, 2019, and 2020, we found similar results where the period of 5/19–7/26 had a higher ambient concentration of SO_2. This pattern may relate to season variation, but the contributors to increasing ambient SO_2 levels were still unclear, which warrants future exploration.

Although the mean concentrations for most of the VOCs and aldehydes obtained from the ambient air were reduced during the Level 3 warning (Table S5), there was no statistical power to test for significant differences between the two restrictive measures due to the small sample sizes (n = 4). Ambient mean concentrations of toluene decreased by 27 % during the Level 3 warning, compared with the Level 2 alert (Table S5). The difference in personal exposure to toluene between the two periods was negligible (Table 2). Ambient toluene in cities is derived mainly from gasoline and automotive emissions; however, several human activities and sources (such as solvent cleaning, printing and publishing, solid waste incineration, and surface coating operations) are also relevant, which likely accounts for the negligible reduction in personal exposures to toluene during the Level 3 warning. Previous studies have reported that the concentrations of ambient VOCs were lower during lockdown periods than during pre-COVID-19 periods, due to remarkable reductions in industrial and traffic emissions (Pei et al., 2022; Zhang et al., 2022). For instance, Pakkattil et al. (2021) reported an overall decrease in benzene, toluene, ethylbenzene, and xylene of 80 %, 75 %, 88 %, and 80 %, respectively, in the Indian metro cities during the COVID-19 lockdown when compared with the pre-lockdown period. Jia et al. (2020) revealed that ambient VOC levels in the Yangtze river delta region of China were reduced by 40 % to 50 % from the non-control period to the control period of the COVID-19 outbreak. Pei et al. (2022) also reported a 19 % reduction in ambient VOC levels in the Pearl River Delta region of south China during the COVID-19 lockdown period, where alkanes, alkenes, and aromatics decreased by 19.0 %, 24.8 %, and 38.2 %, respectively. Satellite remote sensing showed that Wuhan experienced a significant reduction in formaldehyde by 11 % during the COVID-19 outbreak (Ghahremanloo et al., 2021). Additionally, Ninyà et al. (2022) conducted a study of the impact of anti-COVID-19 measures on indoor and outdoor VOC concentrations (including aldehydes) in schools and houses in Tarragona, Spain; they showed that the indoor concentrations were three times higher than those in 2019 because cleaning products and hydroalcoholic gels were important emission sources. In the same study, outdoor VOCs were more strictly associated with combustion processes in automobile traffic and industrial activities. The remarkable reductions in ambient VOCs and aldehydes with strict control measures of COVID-19 published in previous studies may significantly reduce potential health risks of cancer and non-cancer in the population.

Although the strictest control measure of Level 4 (lockdown) was not implemented in Taiwan, the abatement of VOC concentrations measured in the personal exposure study for the Level 3 warning was noticeable. Note that the subjects were exposed to much higher concentrations of VOCs and aldehydes than the ambient levels (Table 3 vs. Table S5), This indicates that ambient measurements may underestimate the impact of anti-COVID-19 measures on personal exposure to highly toxic compounds, including benzene, 1,3-butadiene, and formaldehyde. Personal exposure to specific VOCs (such as benzene, ethylbenzene, o-Xylene, and 1,3-butadiene) associated with on-road vehicle emissions were significantly reduced,

Table 3
The ILCR and HI calculated from inhalation exposure to VOCs and aldehydes during the periods of Level 2 alert and Level 3 warning.

Pollutants	Total ILCR					HI				
	Level 2 alert		Level 3 warning		*p*-Value*	Level 2 alert		Level 3 warning		*p*-Value*
	AM	95th	AM	95th		AM	95th	AM	95th	
VOCs	7.12×10^{-5}	3.95×10^{-4}	4.14×10^{-5}	7.54×10^{-5}	0.039	1.33	3.13	0.668	1.55	<0.001
Aldehydes	2.34×10^{-4}	5.36×10^{-4}	3.08×10^{-4}	5.46×10^{-4}	0.008	3.58	8.48	4.68	8.92	0.008

Total ILCR: sum of all cancer risks calculated from individual VOCs, AM: arithmetic mean, P95: 95th percentile.
* Student *t*-test.

by ~40 to 80 %, compared with the Level 2 alert. The reductions confirm this decrease in traffic-related VOCs in traffic flow and MRT volume within the study area during the sampling period (Table S10). The PMF results also showed that the contribution of vehicle exhausts to ambient VOCs and aldehydes was reduced to 27.8 % (from 33.7 %) from the Level 2 alert to the Level 3 warning (Figs. S1 and S2); here, the high percentage of MTBE, benzene, toluene ethylbenzene, o-xylene, and 1,3-butadiene in Factor 1 associated with traffic-related emissions from gasoline and diesel engine vehicles has been reported (Alves et al., 2015; Hsieh et al., 2011). The emission characteristics of traffic-related VOCs are not only related to the volume of on-road vehicles but also traffic composition, i.e., counts of motorcycles, passenger cars, buses, utility vehicles, and heavy-duty trucks. Although the counts of on-road vehicles (cars, motorbikes, buses, and trucks) showed significant differences between the two restrictive levels, we were not able to distinguish the impact of vehicle types on the characteristics of VOCs and aldehydes obtained from the personal samples.

We cannot rule out the effects of the temporal and seasonal variations on personal exposure to selected VOCs and aldehydes during the study periods. Ambient air quality is strongly affected by meteorological conditions which may explain observed variations in pollutant concentrations (He et al., 2017; Liu et al., 2020; Markowicz and Larsson, 2015; Song et al., 2019; Tsai et al., 2021). For example, Justo Alonso et al. (2022) reported significant seasonal differences in formaldehyde concentrations during the COVID-19 pandemic in Norway, and an increase in temperature may promote the increase in biogenic and anthropogenic emissions and oxidation processes from gaseous air pollutants and precursors. The elevated exposure to formaldehyde and other aldehydes may also associate with personal activities. For instance, the participants with high exposure to formaldehyde and propionaldehyde when reported higher percentages of their time at home and cooking during the Level 3 warning compared to the Level 2 alert. However, the results above did not reflect the effects of personal exposure in the model (Table S8). The response to yes/no questions associated with personal activity information from subjects may not fully capture personal exposure to selected air pollutants. For instance, much higher concentrations of formaldehyde emitted from building materials and furniture indoors than outdoors were mainly reported (Ninyà et al., 2022; Poulhet et al., 2014; Trocquet et al., 2021). In addition, formaldehyde is used as a disinfectant and sterilant formulation to kill fungi, spores, bacteria, and viruses at appropriate concentrations (Ghafoor et al., 2021), and was likely more widely used to reduce the SARS-CoV-2 virus. This was despite 75 % of the market's medicinal alcohol being in shortage during the Level 3 warning in Taiwan. Identifying types of indoor microenvironments, building materials, and furniture might be more effective for exposure predictions. As the predominant sources of ambient formaldehyde in urban areas, vehicle emissions may have partially contributed to personal exposure during the period spent outside, although ambient concentrations of formaldehyde between two restrictive levels were similar (Table S5). As a result, Factor 2 contributed by a high percentage of aldehyde compounds, was referred to as mixed sources and secondary reactions.

In addition to traffic sources, indoor activity and solvent use may contribute to personal exposure to selected VOCs and aldehydes, where chloroform, chlorobenzene, and acrylonitrile with a high proportion in Factor 3 for both periods were identified (Figs. S1 and S2). The percentage of indoor activity and solvent use sources contributing to personal VOC and aldehyde exposures was increased to 34.7 % (from 21.4 % for Level 2 alert) for the Level 3 warning. However, opposite results on chloroform, chlorobenzene, and propionaldehyde with high exposure were obtained when the subjects reported spending more time indoors (Tables S7 and S8). In this study, the variables of personal activities (i.e., cooking, cosmetic use, and indoor time) did not explain the concentrations of exposure to selected VOCs and aldehydes in the GEE model. The reasons likely derived from the design of the questionnaire, limited sample sizes, and complicated within-subject activities and exposure sources. For instance, our integrated sampling for individual exposure to VOCs and aldehydes on a day did not reflect the daily within-subjects activities associated with various activities, sources, and microenvironments.

It should be noted that aldehyde-driven cancer and non-cancer risks for the study population exceeded unacceptable levels (ILCR > 10^{-4} and IH > 1). Across all the datasets, exposure to formaldehyde accounted for 90 % of the total ILCR and 50 % of the HI. Notably, acrolein exposure accounted for 50 % of the HI, which should not be ignored, although exposure concentrations between Levels 2 and 3 were not significantly different in the adjusted GEE model (Fig. 2).

5. Conclusion

Due to unprecedented prevention efforts during the COVID-19 pandemic in Taiwan (Summers et al., 2020), we were unable to evaluate the potential impact of the strictest level of control (i.e., lockdown, Level 4) on air quality and personal exposure to air pollutants. However, compared with the Level 2 alert period, we found that measures under the Level 3 significantly reduced air pollutants (such as CH_4, THC, CO, and NO_2) and personal exposure to specific VOCs (such as benzene, MTBE, xylene, ethylbenzene, and 1,3-butadiene). This was primarily reflected the reductions in transportation (i.e., daily volume of MRT passengers and on-road vehicles). It is notable that personal exposure to formaldehyde and the resultant health risks increased during the period of stricter control measures, which was linked to the period of time spent indoors and the use of disinfectants. Overall, the magnitude of the control measures implemented during the COVID-19 Level 2 alert and Level 3 warning periods have allowed us to improve our understanding of the effect of personal and societal behaviors on exposure to specific VOCs and aldehydes and the associated health risks.

CRediT authorship contribution statement

Yu-Chuan Yen: Writing – original draft, Application of statistical; Chun-Hung Ku: data collection and chemical analyses, experiments; Ta-Chih Hsiao: Investigation, computational; Kai-Hsien Chi: Investigation; Chiung-Yu Peng: Chemical analyses; Yu-Cheng Chen: Conceptualization, Supervision, Visualization, and Project administration.

Data availability

Data will be made available on request.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.scitotenv.2023.163275.

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