



An Environmental Chamber Study of the Characteristics of Air Pollutants Released from Environmental Tobacco Smoke

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ABSTRACT

Environment tobacco smoke (ETS) is an important source of anthropogenic pollution in indoor environment. This paper reports an environmental chamber study of pollutants released from ETS generated by smoking cigarettes in the chamber. Six cigarettes samples sold in Hong Kong and the Mainland of China were characterized. Gaseous pollutants: carbon monoxide (CO), sulphur dioxide (SO₂), nitric oxide (NO), nitrogen dioxide (NO₂), methane (CH₄), non-methane hydrocarbon (NMHC), carbonyls and volatile organic compounds (VOCs); and particulate matter (PM) including organic carbon (OC), elemental carbon (EC) and total carbon (TC) were determined using online and offline analytical methods during smoking and post-smoking periods. Acetaldehyde, acetone and formaldehyde were the three most abundant carbonyls. A total of 18 aromatic and chlorinated VOCs were quantified. Among them, benzene and toluene were the two most abundant VOCs. OC was more dominant (> 93% of TC) than EC. The amounts of tar and nicotine in the cigarettes could have a direct correlation to the PM emitted. Menthol, an additive in cigarettes could also contribute to the ETS pollutants. The indoor ETS could be removed by a higher air exchange rate that would also minimise secondary VOCs formation.

Keywords: Environmental tobacco smoke; Chamber; Emission factors; VOCs; PM_{2.5}.

INTRODUCTION

Environmental tobacco smoke (ETS) consists of a complex mixture of gaseous and particulate pollutants produced from the sidestream and diluted exhaled mainstream smoke from the combustion of tobacco products (Guerin *et al.*, 1992). Sidestream tobacco smoke is defined as the undiluted plume generated from the smouldering end of a cigarette, and the mainstream smoke is the undiluted puff of smoke that is drawn through the cigarette and then exhaled by a smoker (Klepeis *et al.*, 2003).

ETS is classified as Group A carcinogen (U.S.EPA, 1994). The World Health Organisation (WHO) and United States Environmental Protection Agency (U.S.EPA) have published research reports confirming that ETS could increase

the risks of lung cancer, heart disease, and respiratory tract infections (U.S.EPA, 1992; WHO, 1999). ETS is also regarded as an important indoor pollution source (Jones, 1999; Edwards *et al.*, 2001). The majority of the deaths of which ETS is implicated; ETS has been associated with deaths by heart disease (65%) followed by lung cancer (8%) (U.S.EPA, 1992). The International Agency for Research on Cancer (IARC) (IACR, 2002) estimated that the proportion of lung cancer cases that could be attributable to cigarette smoking has reached 90% of the reported populations who are known to be long term cigarette smokers. Other diseases that have been associated with long-term exposure to ETS include increased risk of illness due to strokes, colon polyps and cancers of various organs, including: nasopharynx, oesophagus, larynx, throat, bladder and colon (U.S.EPA, 1992; IACR, 2002). Short-term exposure of ETS could cause irritation to eyes, nasal passages and respiratory system. The effect on the respiratory system could lead to increased wheezing and coughing; and could provoke asthma attacks to susceptible people (Al Frayh *et al.*, 2001; Nguyen *et al.*, 2001).

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Some air contaminants released due to tobacco smoke such as formaldehyde, acetaldehyde, acrolein, polycyclic aromatic hydrocarbons (PAHs), and nitrosamines could also have carcinogenic effects on people (Lofroth *et al.*, 1991; Li *et al.*, 1993; Chao *et al.*, 1998; Jenkins *et al.*, 2000; Singer *et al.*, 2003; Culea *et al.*, 2005a, b; Gee *et al.*, 2005a, b; Holcatova *et al.*, 2005; Vainiotalo *et al.*, 2008; Gu *et al.*, 2010; Lai *et al.*, 2010; Zhu *et al.*, 2010; Hwang *et al.*, 2011). Their emission factors have been determined in many studies (Daisey *et al.*, 1998; Bi *et al.*, 2005). More than 4,000 compounds including toxic volatile organic compounds (VOCs) have been identified in ETS (Daisey *et al.*, 1998). Formaldehyde and acetaldehyde were found to produce the highest emission factors. The estimated ETS emission factor for formaldehyde was 1.3 ± 0.3 mg/cigarette smoked; and for acetaldehyde, 2.2 ± 0.5 mg/cigarette smoked. The emission factor for benzene was estimated to be 406 ± 71 μ g/cigarette smoked. Bi *et al.* (2005) also reported that the average emission factors for total VOCs (TVOC) and total carbonyls were 2.4 ± 0.6 and 4.3 ± 0.6 mg/cigarette smoked, respectively. ETS can also greatly affect the airborne particulate matter (PM) level in indoor environments. Spengler *et al.* (1981) found that the mean PM₁₀ concentrations in 35 homes with non-smokers was 24.4 μ g/m³; but in 15 homes with one smoker in each home, the average PM₁₀ concentration was 36.5 μ g/m³. In the 5 homes surveyed where two of the occupants were smokers, the average PM₁₀ concentration was shown to be as high as 70.4 μ g/m³. The PM_{2.5} emission factors could range from 10 to 20 mg/cigarette smoked (Hildemann *et al.*, 1991). Klepeis *et al.* (2003) investigated size-specific emission factors for ETS particles and found that ETS could yield an average mass median diameter of 0.3 μ m and the total particle emission rate was 0.2–0.7 mg/min per cigar smoked and 0.7–0.9 mg/min per cigarette smoked.

The objectives of this paper were to determine the indoor levels of pollutants, generated by smoking cigarettes in an environmental chamber. The pollutants determined were: carbon monoxide (CO), sulphur dioxide (SO₂), nitric oxide (NO), nitrogen dioxide (NO₂), methane (CH₄), non-methane hydrocarbons (NMHC), carbonyls, and PM_{2.5}; and particulate matters containing organic carbon (OC), elemental carbon (EC) and total carbon (TC). The determination was related to the ETS generated from cigarettes sold in Hong Kong and the Mainland of China and to calculate exposure-relevant emission factors for the different pollutants generated by smoking the different types of cigarettes. To our best knowledge, such emission testing of ETS generated by smoking the local brand of cigarettes has never been performed or reported in literature.

METHODS

Cigarette Samples

Six commercial brands of cigarettes were selected for the smoking emission tests. These are among the top selling brands available on the markets in Hong Kong and Mainland China. Their general information is shown in Table 1. All cigarette samples were fitted with cellulose acetate filters, approximately 20–30 mm in length. Cig A and Cig B are classified as low-tar/nicotine mentholated cigarettes while Cig C to Cig F are regular cigarettes (with tar > 10 mg/cigarette and nicotine \geq 1 mg/cigarette). No information about other additives (e.g., their species and masses) in the cigarettes is given on the packages.

Chamber Experiments

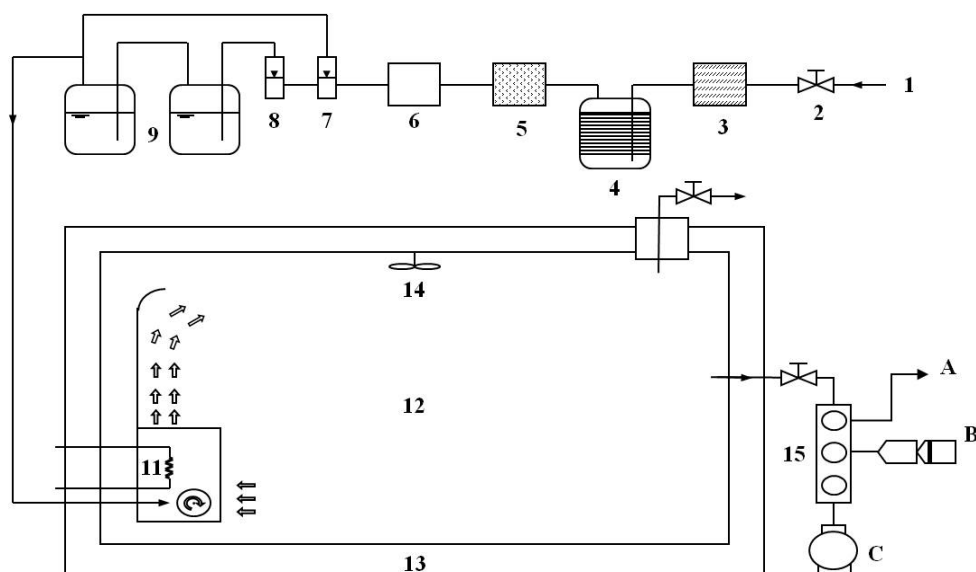
The smoking time for each cigarette was uniform for each cigarette which was approximately six minutes. In each emission test, five cigarettes of the same brand were sequentially smoked in an 18.26 m³ stainless steel environmental test chamber. The total smoking time was 30 minutes for each test. The temperature, relative humidity (RH) of the environmental chamber were maintained at $23 \pm 0.5^\circ\text{C}$ and $50 \pm 5\%$ to simulate the typical indoor air conditions. A TSI portable Q-Trak (model number 8550, TSI Instruments Inc., Shoreview, MN) was put inside the chamber to monitor and tracking of the temperature and RH and that the conditions were within the required ranges.

Fig. 1 shows a schematic diagram of the environmental chamber set-up. The 18.26 m³ chamber was purged by conditioned blower air, which was cleaned and conditioned by passing through an air cleaning system consisting of activated charcoal particle filters and High-Efficiency Particulate Air (HEPA) filters. The temperature of inlet air was controlled by conditioning coils. The relative humidity is controlled by adding appropriate amount of deionized water into the air system. Air exchange rate (ACH) was maintained at 0.5 1/h for the emission tests of smoking Cig A to Cig F. An additional ACH of 2.8 1/h was used for testing the smoking of Cig F in order to evaluate the effect of ACH on the emission factors. Mixing fans were installed at the ceiling of the chamber to ensure adequate air mixing. Before each smoking emission test, the chamber wall was cleaned with water and any remnant of ETS in the chamber air and surfaces were ozonized with ozone oxidation and then conditioned for at least four hours at the set temperature, RH and ACH, prior to the start of another testing.

A sampling port with Teflon tubing was inserted into the centre of the chamber at 0.6 m above floor level. The sample

Table 1. A summary of cigarette samples in the environmental chamber study.

Cigarette Sample id	Country of origin	Tar content (mg)	Nicotine content (mg)	Specification
Cig A	USA	1.0	0.1	Mentholated
Cig B	USA	6.0	0.5	Slim and mentholated
Cig C	USA	12	1.0	Regular
Cig D	USA	12	1.2	Regular
Cig E	Mainland of China	15	1.3	Regular
Cig F	Mainland of China	15	1.0	Regular



Indication:

1. Air inlet, 2. Valve, 3. Blower, 4. Active charcoal filter, 5. HEPA filter, 6. Mass flow controller, 7. Flow controller wet air, 9. Humidifier, 10. Rotating cylinder,
11. Heating unit, 12. Large environmental test chamber, 13. Incubator, 14. Mixing fan,
15. Sampling manifold, 16. Air outlet.
- A. Gas analyzers for criteria gases, B. Ozone scrubber & DNPH cartridge for carbonyls,
- C. Canister for VOCs, D. Real-time $PM_{2.5}$ monitor,
- E. Mini-volume sampler for $PM_{2.5}$ and carbonaceous analysis.

Fig. 1. Schematic diagram of the environmental chamber for the study.

port was about 0.75 m away from the smoked cigarette and the emission from ETS would be fully mixed in the indoor air of the chamber before being drawn by the air sampler. Air samples in the chamber were collected through the sampling port to different samplers or analyzers connected in series. VOCs samples were collected by pre-evacuating SUMMA canister at a rate of 4.0–6.0 L/min using mass flow controllers (model FC4101CV-G, AutoFlow Inc., CA). Carbonyls were collected by drawing air through a cartridge impregnated with acidified 2,4-dinitrophenylhydrazine (DNPH) (Water Corporation, Sep-Pak DNPH-silica, Milford, MA) with a flowrate of 0.8–1.0 L/min (USEPA, 1999). An ozone scrubber was connected before the DNPH-silica cartridge to prevent interference from ozone. A GFC Ambient CO Analyzer (Model 48, Thermo Environmental Instruments Inc.), a Chemiluminescence NO-NO₂-NO_x Analyzer (Model 42C, Thermo Environmental Instruments Inc.) and a Direct Methane, Non-Methane Hydrocarbon Analyzer (Model 55C, Thermal Environmental Instruments Inc.) were also connected and used to quantify CO, carbon dioxide (CO₂), NO_x, CH₄ and NMHC concentrations, respectively. The flow rates of the instrument were measured with a rotameter. The rotameter was calibrated in the laboratory against a soap bubble flow meter.

Sampling and Analytical Methods

Sampling events were divided into three different periods including I) pre-smoking (30 minutes), II) smoking (30 minutes), and III) post-smoking (60 minutes) periods.

Background levels of pollutants in the chamber were determined in the Period I, which were all well below the guidelines given in the Large Chamber Test Protocol for Measuring Emissions of VOCs and Aldehydes suggested by U.S.EPA (U.S.EPA., 1999). The background in the chamber air were kept $< 10 \mu\text{g}/\text{m}^3$ for total volatile organic compounds (TVOCs) and $< 2 \mu\text{g}/\text{m}^3$ for any individual VOC. The smoking emission testing period (II) started when the first cigarette was lit and the fifth cigarette was extinguished which was a total of 30 minutes. Changes in concentrations of air pollutants were further monitored in the post smoking period (Period III).

Concentrations of CO and CO₂ were measured with the CO Analyzer. The time interval for each data point was 1 min. Concentrations of NO, NO₂ and NO_x were measured using the Chemiluminescence NO-NO₂-NO_x Analyzer. The concentrations of CH₄ and total NMHC were measured using the Direct Methane/Non-Methane Hydrocarbon Analyzer. A five-point calibration and a zero check were performed daily for each instrument using certified standard gas of known concentrations (CO in N₂: 500 ppmv, Arkonic Gases and Chemicals, Hong Kong, China; NO_x in N₂: 51.75 ppmv, Airgas, Lenexa, KS; methane/propane in air: 6.3 ppmv/5.6 ppmv, Air Liquide, Paris, France; TO-14 Calibration gas mixture: 1 ppm in N₂, Supelco, Bellefonte, PA). The linearity of the calibration curve was indicated by a correlation of determination (R^2) of at least 0.999. The ranges and method detection limits (MDLs) of the analytical methods are summarized in Table 2. A Dust-Trak air monitor (Model

Table 2. Detection ranges and limits of the sampling methods.

Air parameter	Detectable mechanism	Range	Minimum detection limit
Carbon dioxide (CO ₂)	Non-dispersive infra-red analyzer	0–5000 ppmv	1 ppmv
Carbon monoxide (CO)	Non-dispersive infra-red analyzer	0–50 ppmv	0.1 ppmv
Nitrogen oxide (NO _x)	Chemiluminescence based analyzer	0–1000 ppbv	0.40 ppbv
Methane and non-methane hydrocarbon (NMHC)	Gas chromatography/flame ionization detection (GC/FID)	0–200 ppmv	0.2 ppmv (methane) 0.05 ppmv (NMHC)
VOCs	Gas chromatography/mass spectrometry (GC/MS) USEPA TO-14 Method	0–5000 µg/m ³	0.2 µg/m ³
Carbonyls	High pressure liquid chromatography (HPLC) USEPA TO-11 Method	0–5000 µg/m ³	0.1–0.2 µg/m ³

8520, TSI Instruments Inc.) was used to continuously monitor the PM_{2.5} concentrations during the sampling period I to III. The Dust-Trak air monitor was calibrated against a PM_{2.5} mini-volume sampler which had a linear correlation > 0.9.

Apart from the real-time PM_{2.5} mass monitoring, offline PM_{2.5} samples was collected onto a 47 mm Teflon-membrane and quartz-fibre filters (Whatman, Clifton, NJ), respectively, with two parallel mini-volume samplers, at a flow rate of 5 L/min drawn from the environmental chamber. The mass of PM_{2.5} collected on the Teflon-membrane filters was analyzed by gravimetry. Each filter used for gravitational sampling was conditioned at approximately 50% RH for 24 h before sampling and weighing. The filters were weighed at least three times using an electronic micro-balance (Model A200 S-D1B, Sartorius, Goettingen, Germany). Operation blank samples, which were processed simultaneously with field samples, were collected to access the adsorption of organic components onto the filter during sampling. The aerosols collected on the quartz-fibre filters were quantified and the quantity of OC and EC were measured by thermal optical reflectance (TOR) as given by the IMPROVE protocol for the operation of a DRI Model 2001 Thermal/Optical Carbon Analyzer (Atmoslytic Inc., Calabasas, CA) (Chow and Watson, 2002). The MDL for the carbon analysis was 0.8 and 0.4 µg C/cm² for OC and EC, respectively, with a precision better than 10% for total carbon (TC). It must be noted that few semi-volatile organic compounds (SVOCs) of interest (but not included within the present scope of testing) existed in the vapour phase or associated with PM.

Analytical procedures for the determination of VOCs were based on the U.S.EPA TO-14 method (U.S.EPA., 1998). Ambient volatile organic canister samplers (AVOCS) (Series 97–300, Andersen Instruments Inc., Smyrna, GA) were used to collect air samples into pre-cleaned and pre-evacuated 2-L stainless steel canisters for 1 h during Period I, for 15 minutes twice at time 0 min and time 15 min respectively during Period II, and for 1 hr during period III with a flow rate of 30 mL/min drawn from the environmental chamber. The canisters were pressurized when sampling. The analytical system used to analyze VOCs (i.e., saturated, unsaturated, aromatic, and halogenated hydrocarbons) involved a cryogenic pre-concentration of 1520 ± 1 cm³ (STP) of air sample in a stainless steel tube filled with glass beads (1/8" diameter) and immersed in liquid nitrogen (–196°C). A mass flow controller with a maximum allowed flow of 500 mL/min was used to control the trapping process. The trace

VOCs were revolatilized using a hot water bath and then directed to a gas chromatography/mass spectrometric detector (GC/MSD) system (GC6890/5973 MSD, Hewlett Packard, Wilmington, DE). A total of 18 VOCs were identified and quantified in this study. The identification and quantification of VOCs were based on retention time and peak areas of the corresponding calibration standards, respectively. These tasks were performed by matching spectra using the National Institute of Standards and Technology (NIST) mass spectra library, and also with TO-14 standard calibration gas (Toxi-Mat-14M Certified Standard, Spectra Gases, Branchburg, NJ) based on retention times. Calibration curves for all measured VOCs were prepared and the R² for these calibration curves were greater than 0.95. For the determination of MDLs of the GC/MS system, TO-14 standard calibration gas at 0.2 µg/m³ was analyzed seven times.

Carbonyls were collected by drawing the chamber air through a silica gel cartridge impregnated with acidified DNPH (Waters Sep-Pak DNPH-silica). An ozone scrubber (Waters Corporation) was connected before the DNPH-silica cartridge in order to prevent interference from ozone. Time-integrated air samples were taken from the environmental chamber with a flow rate of approximately 1000 mL/min. The sampling times were 1 h (Period I), 0.5 h (Period II), and finally 1 h (Period III). No breakthrough was found at this range of flow rates and sampling time. Each DNPH cartridge was eluted with 5.0 mL of acetone-free acetonitrile (HPLC/GC grade) to a volumetric flask. Certified calibration standards of 12 DNPH-carbonyl derivatives were purchased from Supelco (Bellefonte, PA) and diluted into concentration ranges of 0.05 to 2.0 µg/mL. The R² of the calibration plot was at least 0.999. The samples and standards were analyzed by injecting 20 µL of the solutions to a high pressure liquid chromatography (HPLC) system (Waters Breeze™ HPLC System equipped with a 1525 binary HPLC pumps), equipped with an ultraviolet (UV) detector. Absorbance at 360 nm was used for the quantification of the DNPH-carbonyl derivatives. The MDL of the target carbonyls was equivalent to 0.1 to 0.2 µg/m³ in chamber air.

Exposure-relevant emission factors (EREFs) for carbonyls and VOCs were calculated using Eq. (1):

$$EREF = \frac{\sum_{pd} (C_{pd} - C_{pre}) \lambda V \Delta t}{M} \quad (1)$$

The concentrations of VOCs and carbonyls measured

during the smoking periods (C_{pd}) were firstly subtracted by the background concentrations (C_{pre}). These net concentrations in $\mu\text{g}/\text{m}^3$ were then multiplied by the air exchange rate λ (1/h), chamber volume V (18.26 m^3), and duration Δt (h) to calculate the mass (μg) of each constituent removed by means of ventilation from the chamber during each smoking period (Singer *et al.*, 2003). The total mass emitted from a source was calculated by adding the mass removed from the chamber by ventilation to the mass remained in the chamber. EREFs were finally calculated by dividing the total mass emitted from a source by the mass of specimen burned M (g).

RESULTS AND DISCUSSION

Criteria Gas Pollutants

The average net concentrations of CO, NO, CH_4 , and total NMHC in the chamber air after deducted from the background concentrations, during and after the smoking of six cigarette samples (Period II and III) are shown in Table 3. The CO concentrations ranged from 7.8 to 14.4 ppmv during the cigarette smoking (Period II). The concentrations produced by smoking the cigarette samples, except Cig B, exceeded the “Good Classes” criteria of 8.7 ppmv specified by Hong Kong Environmental Protection Department (HKEPD) as Indoor Air Quality Objectives (IAQO) for office buildings and public places (HKEPD, 2003). Smoking Cig D, produced the highest concentration of NO (285 ppbv) which was more than 2.5 times higher than the lowest concentration produced when smoking Cig F (100 ppbv). The concentrations of CH_4 and total NMHC in the chamber air during smoking (period II) were in a range of 0.8 to 2.0 ppmv and 1.2 to 2.1 ppmv, respectively.

Smoking cigarettes with low-tar/nicotine contents (Cig A and B) produced the lowest CH_4 concentrations (0.8 ppmv), compared to an average of 1.5 ppmv when smoking the regular brand cigarettes (Cig C-F). The findings illustrate that the tar and nicotine burning could significantly contribute to indoor CH_4 concentrations. Similar trend was found for the other criteria gases. The average CO and total NMHC concentrations produced when smoking the cigarettes with low-tar/nicotine content were 9.8 and 1.4 ppmv, respectively, which were again lower than the average concentrations of 12.5 and 1.8 ppmv, respectively, produced when smoking the regular cigarettes. Even though there was only a ppbv-level increase in NO concentration due to the cigarettes smoking, the emission was still a positive association with the tar and nicotine contents in the cigarettes.

Clearly, the average concentrations of the criteria gases (CO, NO, and CH_4) would decrease after the smoking had stopped (Period III) (Fig. 2). However, higher average total NMHC concentrations were found in the post-smoking time-integrated (period III) air samples; this was possibly due to the reformations or re-emissions of NMHCs from PMs which were absorbed onto the chamber surfaces during the smoking period (period II). The concentrations of SO_2 and NO_2 in the chamber air during smoking were below MDL, suggesting ETS is not the pollution source for these compounds.

Carbonyl Compounds

Tobacco smoking is one of the major contamination sources of carbonyls in indoor environment (Hodgson *et al.*, 1996; Marchand *et al.*, 2006; Bari *et al.*, 2011; Panagopoulos *et al.*, 2011). The average concentrations of 12 carbonyl compounds measured from the time integrated samples collected from the chamber air during smoking (Period II) and post-smoking periods (Period III) are shown in Table 3. Acetaldehyde was the most abundant carbonyls produced by smoking cigarettes which averagely accounted for 45% of the total carbonyls quantified. Acetone and formaldehyde were the next two most abundant carbonyls produced by smoking the cigarette samples, which averagely accounted for 16%, and 14%, respectively of the total carbonyls. Feng *et al.* (2004) measured 21 carbonyls in four residential hotel ballrooms, demonstrating that cigarette smoking would be a major source of indoor carbonyls. Our result is consistent with their findings that acetaldehyde was the most abundant carbonyls, accounting for 51% of the total indoor carbonyls, followed by formaldehyde, 22%. Fenske and Paulson (1999) suggested that human breath could be an emission source of VOCs, especially acetone.

No association was apparent between the amounts of tar and nicotine contents in cigarettes and the carbonyls concentrations produced by smoking during the chamber tests. Mentholated Cig A has the lowest tar and nicotine contents (1.0 and 0.1 mg/cigarette respectively); however, it produced the highest concentrations of acetaldehyde, acetone, acrolein, propionaldehyde, crotonaldehyde, 2-butanone, butyraldehyde, benzaldehyde, valeraldehyde, and *m*-tolualdehyde during smoking in comparison to all the other cigarette samples when smoked during test. In contrast, mentholated Cig B is another low tar- and nicotine-containing cigarettes (6 mg and 0.5 mg/cigarette respectively) and smoking this cigarette produced the lowest concentrations of acetaldehyde and 2-butanone. The data demonstrate that other materials, e.g., additives in the cigarettes, could greatly control the formations of carbonyl. However, no further information is given regarding the additives content in the cigarettes. Interpretation to this effect could not be confirmed by the data.

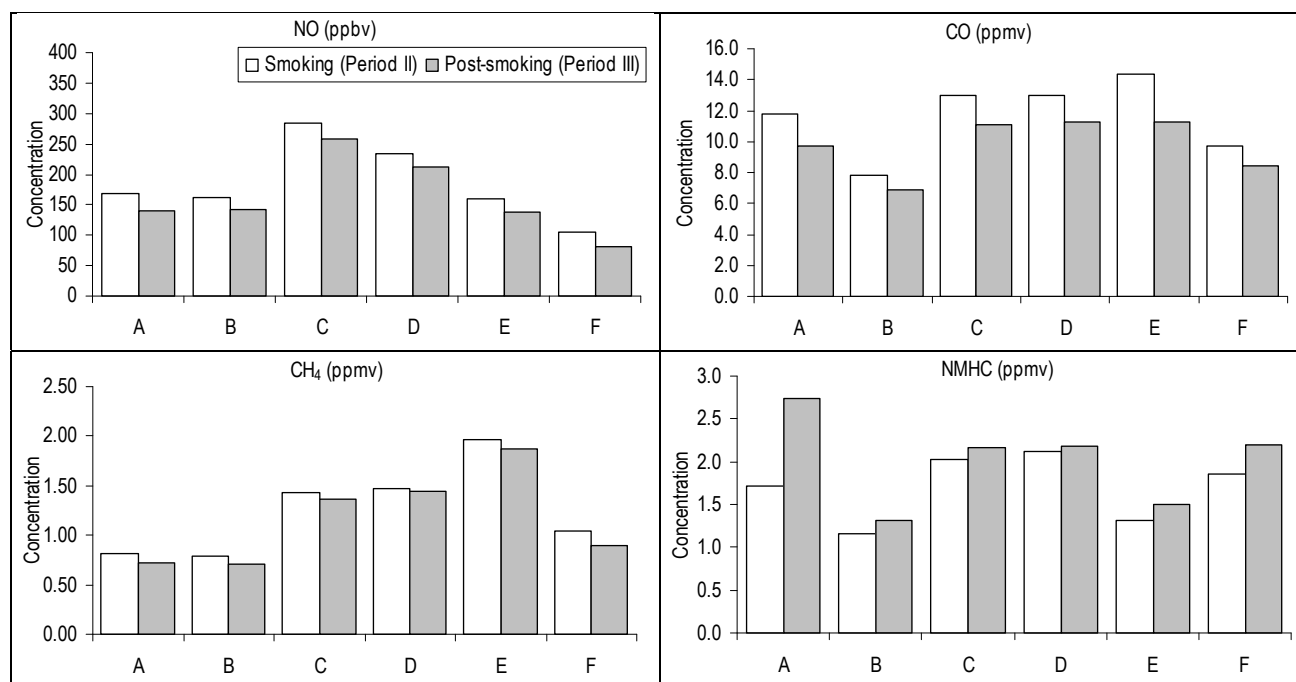
Formaldehyde is a known human carcinogen and could cause eye or respiratory tract irritations at ppbv levels (IACR, 1995; Yu and Kim, 2010). According to the Hong Kong IAQO, the indoor formaldehyde concentration should be limited to 100 $\mu\text{g}/\text{m}^3$. The emission testing of cigarette smoking showed that all cigarettes except for Cig D produced concentrations exceeded this criterion.

The calculated emission factors of the carbonyls in unit of $\mu\text{g}/\text{cigarette}$ are shown in Table 4. The amounts of carbonyls emitted due to smoking of the six cigarette samples ranged from 370 to 790 $\mu\text{g}/\text{cigarette}$ for formaldehyde; 1,900 to 4,600 $\mu\text{g}/\text{cigarette}$ for acetaldehyde; and 310 to 1700 $\mu\text{g}/\text{cigarette}$ for acetone. Daisey *et al.* (1998) determined the emission factors for 21 VOCs produced by smoking cigarettes in a room-sized environmental chamber. They reported that smoking had produced highest emission factors for acetaldehyde and formaldehyde among all the quantified compounds, which were $2,200 \pm 500$ $\mu\text{g}/\text{cigarette}$ and

Table 3. Concentrations of gaseous pollutants during smoking emission and post-smoking period.

	Cigarette Samples											
	A			B			C			D		
	smoking ^a	post-smoking ^b	smoking	post-smoking	smoking	post-smoking	smoking	post-smoking	smoking	post-smoking	smoking	post-smoking
CO (ppmv)	11.8	9.7	7.8	6.9	13.0	11.1	13.0	11.3	14.4	11.3	9.7	8.4
NO (ppbv)	168.1	140.9	160.7	142.0	284.4	256.9	234.7	211.1	159.0	138.0	106.0	81.2
CH ₄ (ppmv)	0.8	0.7	0.8	0.7	1.4	1.4	1.5	1.4	2.0	1.9	1.0	0.9
NMHC (ppmv)	1.7	2.7	1.2	1.3	2.0	2.2	2.1	2.2	1.3	1.5	1.9	2.2
Carbonyls (µg/m ³)												
Formaldehyde	180	56	130	82	180	95	94	51	210	110	220	95
Acetaldehyde	580	640	240	310	490	750	330	470	460	490	350	280
Acetone	200	190	71	45	220	280	150	200	220	170	100	65
Acrolein	61	120	22	39	51	86	22	75	39	89	18	19
Propionaldehyde	120	15	26	10	44	80	11	55	7.3	19	11	56
Crotonaldehyde	120	27	46	23	37	68	49	58	64	64	17	18
2-Butanone	64	7.1	6.7	17	12	38	25	32	29	13	26	34
Butyraldehyde	96	16	26	13	21	56	34	52	46	49	18	15
Benzaldehyde	25	4.4	7.8	7.0	4.7	11	10	13	17	13	17	14
Valeraldehyde	30	3.4	14	9.4	bd ^c	bd	6.3	38	23	26	bd	bd
<i>m</i> -Tolualdehyde	24	bd	1.6	3.1	2.0	3.2	bd	bd	4.0	3.3	bd	bd
Hexaldehyde	44	5.7	26	10	28	17	7.4	13	58	29	56	30
Chlorinated compounds (µg/m ³)												
Methyl chloride	3.7	2.5	3.1	5.8	5.3	7.0	8.4	9.9	8.4	8.7	9.0	11
Chloroethene	24	20	15	10	27	22	20	20	28	20	21	19
Ethyl chloride	4.0	2.6	4.0	2.2	7.4	5.2	1.6	0.9	4.5	1.7	8.2	5.4
Methylene chloride	11	17	19	34	2.8	4.9	14	23	15	34	18	29
1,1-Dichloroethane	17	9.1	13	8.6	12	7.9	26	17	17	10	22	16
<i>cis</i> -1,2-Dichloroethene	28	22	60	48	14	12	29	23	45	34	20	15
Chloroform	28	25	16	12	20	12	13	8.6	14	8.9	13	10
1,2-Dichloroethane	31	31	11	9.0	39	36	28	25	29	28	20	19
1,2-Dichloropropane	3.7	4.1	5.7	6.7	4.3	4.6	6.4	7.9	3.5	5.0	5.0	6.3
1,1,2-Trichloroethane	1.8	48	2.3	40	1.5	39	1.8	23	1.3	57	2.6	13
1,1,2,2-Tetrachloroethane	8.7	9.7	6.4	7.2	6.9	8.4	7.1	9.1	8.8	10	5.5	7.3
Aromatics (µg/m ³)												
Benzene	65	59	26	24	80	67	63	56	81	72	76	69
Toluene	120	140	36	49	48	52	50	64	98	120	51	65
Ethylbenzene	16	12	11	7.0	20	15	17	13	17	11	14	11
<i>o</i> -Xylene	4.3	4.1	3.2	1.9	5.9	4.7	4.7	4.1	5.0	4.3	5.1	4.7
<i>m,p</i> -Xylene	22	17	12	7.3	29	23	24	20	20	15	20	16
Styrene	20	18	8.5	6.7	25	20	19	15	18	18	17	14
1,3,5-Trimethylbenzene	9.3	7.8	7.6	5.6	11	7.4	9.1	8.0	9.6	7.8	11	9.1

^a Samples were collected for 30 minutes during ignition and completion of smoking.^b Samples were collected for 60 minutes after smoking.^c bd represent below minimum detection limit.



Remarks:

Smoking (Period II): samples were collected for 30 minutes during ignition and completion of smoking.

Post-smoking (Period III): samples were collected for 60 minutes after smoking.

Fig. 2. Concentrations of criteria gas pollutants during smoking emission and post-smoking periods.

$1,300 \pm 300$ $\mu\text{g}/\text{cigarette}$, respectively. The average emission rate for acetaldehyde produced by the present environmental chamber testing is closely consistent with their reported value but approximately 50% lower for formaldehyde. As the tested samples were from different origins, the variations of emission rates are thus reasonable.

VOCs

Table 3 shows the average concentrations of 18 VOCs found in the time integrated chamber air samples collected during smoking (Period II) and post-smoking periods (Period III). For Cig A, B, and E, toluene was the most abundant VOCs produced by smoking, followed by benzene. A reverse trend was observed for Cig C, D, and F in which benzene was the most abundant VOCs produced by smoking, followed by toluene. The concentrations of toluene and benzene in chamber air ranged from 36 to 120 $\mu\text{g}/\text{m}^3$ and 26 to 81 $\mu\text{g}/\text{m}^3$ respectively during the smoking periods.

No association was found between the toluene emission and the amount of tar, nicotine, and menthol in the cigarettes. Mentholated Cig A had the lowest tar and nicotine content, but produced the highest toluene concentrations of toluene of 120 $\mu\text{g}/\text{m}^3$ by smoking compared with smoking of the other cigarette samples. However, other low-tar/nicotine mentholated Cig B produced the lowest toluene concentration (36 $\mu\text{g}/\text{m}^3$) by smoking during the chamber test. The toluene emission is thus expected to vary due to the different types and amounts of other additives included in the cigarettes.

Emissions of benzene by cigarette smoking showed positive association with the cigarettes' tar and nicotine content. The lowest benzene concentrations were produced

by smoking Cig B (26 $\mu\text{g}/\text{m}^3$). The benzene concentration produced by smoking Cig A (65 $\mu\text{g}/\text{m}^3$) was also lower than those produced by smoking the regular cigarettes with higher tar and nicotine contents (75 $\mu\text{g}/\text{m}^3$ on average). The mass ratios of toluene to benzene produced were 1.83 and 1.35 by smoking mentholated Cig A and B respectively, which were much higher than the values produced by smoking the non-mentholated cigarettes, ranging from 0.60–1.21. Our results were consistent with the findings reported by Heavner *et al.* (1995) that benzene and styrene were not well correlated in ETS. The chemistry of VOCs formation from smoking the two types of cigarettes would be potentially different. However, there is a lack of any proposed mechanism to explain how the transformation of tar, nicotine, menthol, or other additives during smoking of cigarettes had any effect on the emissions of VOCs. The levels of benzene and toluene recommended by the "Good Class" of IAQO of Hong Kong are 16.1 and 1,092 $\mu\text{g}/\text{m}^3$ respectively. The benzene concentrations produced in the chamber air, either during smoking of the six cigarettes (Period II) or post-smoking (Period III), had exceeded the Hong Kong IAQO. The toluene concentrations produced however, were much lower than the IAQO level. The concentrations of other aromatic VOCs (e.g., styrene, o,m,p-xylene) were all below 30 $\mu\text{g}/\text{m}^3$ during smoking in the chamber test period.

Rather than the aromatic VOCs, the chlorinated VOCs were significantly produced by smoking (Table 3). Chloroethane, cis-1,2-dichloroethene, chloroform, and 1,2-dichloropropane, and 1,1,2-trichloroethane were the most abundant chlorinated VOCs found in the chamber air

Table 4. Emission factors of carbonyls and VOCs ($\mu\text{g}/\text{cigarette}$) for the cigarette samples.

	Cigarette Samples					
	A	B	C	D	E	F
Tar (mg/cigarette)	1.0	6.0	12	12	15	15
Nicotine (mg/cigarette)	0.1	0.5	1.0	1.2	1.3	1.0
Carbonyls						
Formaldehyde	470	570	690	370	790	720
Acetaldehyde	4000	1900	4600	2900	3100	1900
Acetone	1200	310	1700	1200	1100	450
Acrolein	710	230	520	430	520	120
Propionaldehyde	190	79	480	310	110	320
Crotonaldehyde	260	170	410	360	410	110
2-Butanone	97	99	220	200	98	210
Butyraldehyde	180	95	330	320	310	99
Benzaldehyde	47	45	65	80	87	92
Valeraldehyde	46	64	bd	210	160	bd
m-Tolualdehyde	bd ^a	18	19	bd	22	bd
Hexaldehyde	71	79	120	78	210	220
Chlorinated compounds						
Methyl chloride	16	27	35	51	47	57
Chloroethene	120	64	130	110	120	110
Ethyl chloride	17	15	33	6.2	14	35
Methylene chloride	82	160	23	110	150	140
1,1-Dichloroethane	64	55	51	110	68	99
cis-1,2-Dichloroethene	130	290	69	140	210	91
Chloroform	140	73	80	55	58	60
1,2-Dichloroethane	170	53	200	140	160	110
1,2-Dichloropropane	22	35	25	41	25	32
1,1,2-Trichloroethane	180	150	150	87	210	52
1,1,2,2-Tetrachloroethane	51	38	43	46	53	37
Aromatics						
Benzene	330	140	390	320	410	390
Toluene	730	250	280	330	620	330
Ethylbenzene	73	46	91	79	71	66
o-Xylene	23	13	28	24	25	26
m,p-Xylene	100	49	140	120	91	95
Styrene	100	40	120	89	99	82
1,3,5-Trimethylbenzene	45	34	47	46	46	53

^a “bd” represents below minimum detection limit.

samples. Unfortunately, formation mechanisms of these compounds are not known. In general, no strong association was found between chlorinated VOCs emission by smoking and the menthol, tar and nicotine contents in the cigarettes, even though the slim-design Cig B, which is the lightest in weight, produced the lowest concentrations of chloroethene and 1,2-dichloroethane by smoking in comparison to smoking of the other cigarettes.

The average concentrations of five VOCs (including toluene, methylene chloride, 1,2-dichloropropane, 1,1,2-trichloroethane, and 1,1,2,2-tetrachloroethane) determined during the post-smoking period were higher than during the smoking period. Among these compounds, a greater increase of 1,1,2-trichloroethane was found during the post-smoking period, ranging from 5 to 43 times higher than during the smoking period. The results demonstrate that, during the post-smoking period, certain VOCs could be reformed; or re-emitted from PM which had been absorbed

onto the chamber surfaces. Table 4 shows the individual emission factors of 18 VOCs produced by smoking the cigarette samples. Bi *et al.* (2005) reported that the average emission factors for total VOCs were 2.4 ± 0.6 mg/cigarette smoked. Our results were generally lower than their findings but the numbers of VOCs quantified were different in the two studies. Daisey *et al.* (1998) reported that the benzene emission factor was 406 ± 71 $\mu\text{g}/\text{cigarette}$, which was close to or slightly higher than our values ranging from 140 to 410 $\mu\text{g}/\text{cigarette}$ smoked during the chamber test.

Particulate Matters

Fig. 3 shows the temporal changes of $\text{PM}_{2.5}$ concentrations for the six cigarette samples. The initial $\text{PM}_{2.5}$ concentrations ranged from 0.01 to 0.22 mg/m^3 in the pre-smoking period (Period I). Its concentration sharply increased and reached to the maximum at the end of the smoking period (Period II), and declined during the post-smoking period (Period

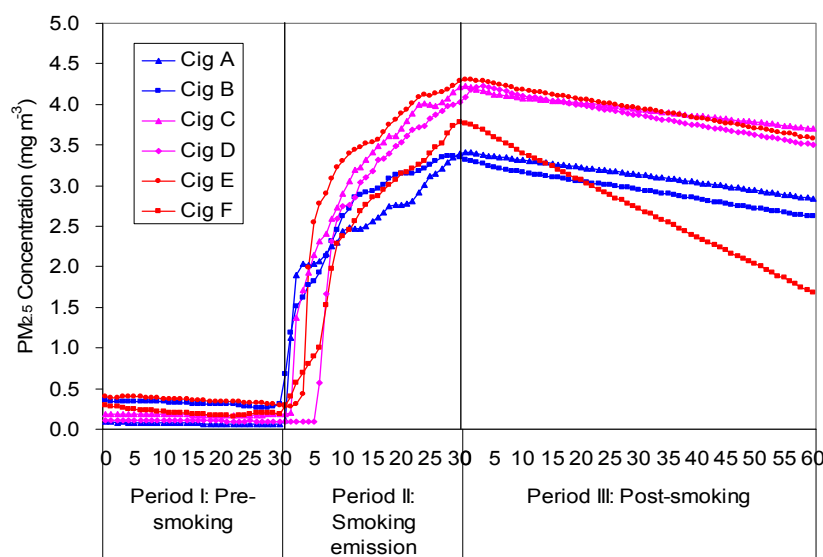


Fig. 3. Temporal variations of $PM_{2.5}$ concentrations in the environmental chamber tests.

III). The emissions of $PM_{2.5}$ showed a positive association with the amounts of tar and nicotine in the cigarettes. Cig E contained the richest tar and nicotine contents and smoking the cigarette produced a maximum $PM_{2.5}$ concentration of 4.22 mg/m^3 ; this was the highest concentration measured as compared to the emissions due to smoking of the other samples. Smoking the low-tar/-nicotine mentholated Cig A and B produced maximum $PM_{2.5}$ concentrations of 3.42 and 3.37 mg/m^3 respectively, which were $\sim 15\%$ lower than the regular cigarettes ($> 4 \text{ mg/m}^3$ except Cig F). Fig. 4 shows a linear relationship between the average $PM_{2.5}$ concentrations measured during smoking and the tar and nicotine contents of the six tested cigarettes. The R^2 for the concentration correlation with tar and concentration with nicotine were 0.65 and 0.86 respectively. The correlations shown by the figure has illustrated that the burning of tar and nicotine during smoking could generate $PM_{2.5}$ into indoor environment.

The decay rates of the $PM_{2.5}$ emitted from the smoking of the regular cigarettes were surprisingly faster than that produced from smoking of the low-tar/nicotine mentholated cigarettes in the environmental chamber of the same ACH condition. Although smoking Cigs C, D, E and F produced highest concentrations of $PM_{2.5}$ but the concentrations declined sharply by $18\text{--}53\%$ during the post-smoking period (Period III). The decline in $PM_{2.5}$ concentrations were much slower after smoking the mentholated Cigs A and B which were only 12% and 11% , respectively as compared to the others. It is possible that the variation in the compositions, rather than menthol content of the cigarettes that had an effect on the variation of the chemical profiles in the particulates produced by smoking the cigarettes.

The results showed that the $PM_{2.5}$ generated from the burning of tar and nicotine by smoking did not have a long lifetime in the chamber atmosphere in the particulate phase under the standard indoor air environment. The PM can persist in the indoor air and could cause impacts on health of building occupants, especially considering the types of chemicals that could be adhere to the particulates generated

from smoking (Singer *et al.*, 2003). So, although smoking the mentholated cigarettes produced the least amount of $PM_{2.5}$, this should still be a source of health risks to people who are immediately exposed to the ETS.

Klepeis *et al.* (2003) investigated the size-specific emission factors for cigarettes smoke particles and reported that cigarettes could yield particles with an average mass median diameter of $0.3 \mu\text{m}$ and the equivalent total particle emission rate of $0.2\text{--}0.7 \text{ mg/min}$ was found for smoking cigars and $0.7\text{--}0.9 \text{ mg/min}$ for smoking cigarettes. Bi *et al.* (2005) reported that the average emission factors for total particulate matters were $15.8 \pm 1.4 \text{ mg/cigarette smoked}$. Our results are consistent with their findings; the emission rate of $PM_{2.5}$ ranged from $0.27\text{--}0.65 \text{ mg/min}$ or $8.5 \pm 2.7 \text{ mg/cigarette smoked}$.

The amount of OC, EC, and TC collected on the quartz-fibre filter samples during smoking (Period II) and post-smoking (Periods III) were determined using offline TOR carbon analysis (Fig. 5). The average OC concentrations ranged from 0.62 to 1.4 mg/m^3 ; EC concentration, from 0.006 to 0.090 mg/m^3 ; and TC, from 0.65 to 1.4 mg/m^3 . EC was not the major particles generated by smoking; and only less than 7% of the TC in the samples. The highest OC, EC, TC concentrations were generated by smoking Cig E, which had the highest tar and nicotine contents. Relatively lower OC and TC concentrations were determined in the emission generated from smoking of the low-tar/nicotine mentholated Cigs A and B.

Inferences from Air Exchange Rate

The effects of ACH on the pollutants concentrations were also investigated in the present environmental chamber study. The same online and offline measurements of gases and PM were conducted during the environmental chamber testing of smoking Cig F at two different ACH; 0.5 1/h (normal) and a higher ventilation rate of 2.8 1/h . The results (Fig. 6) showed that most of the pollutants concentrations were reduced at the higher ACH in comparison to the

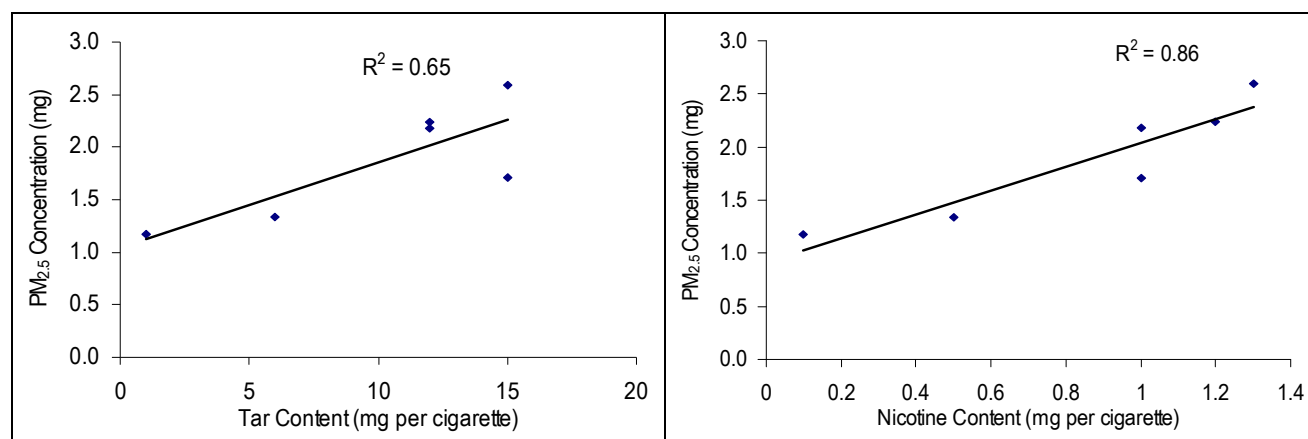
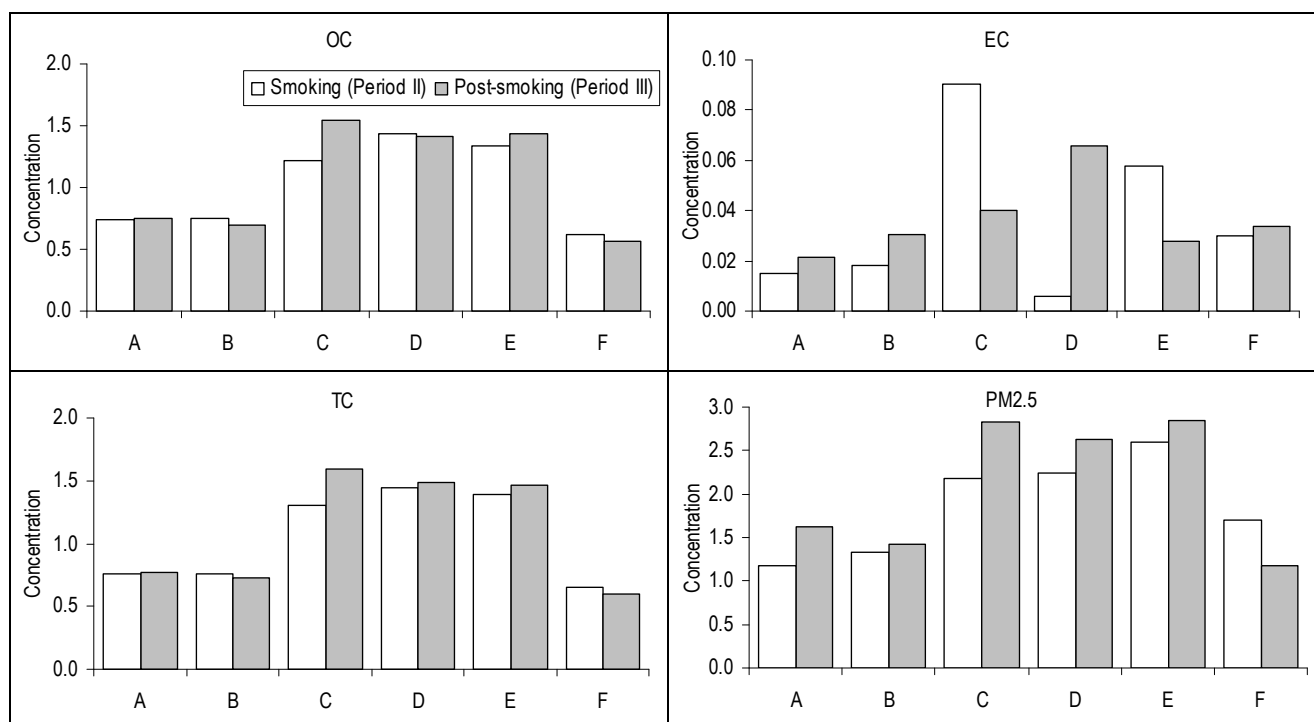


Fig. 4. A relationship plot of average PM_{2.5} concentrations determined in smoking emission period and tar and nicotine contents in the cigarette samples.



Remarks:

Smoking (Period II): samples were collected for 30 minutes during ignition and completion of smoking.

Post-smoking (Period III): samples were collected for 60 minutes after smoking.

Fig. 5. Average concentrations (mg/m³) of OC, EC, TC and PM_{2.5} during smoking emission and post-smoking periods.

normal ventilation; either during smoking (Period II) or the post-smoking (Period III). The enhanced ACH was also shown to be effective for reducing the concentrations of toluene, methylene chloride, 1,2-dichloropropane, 1,1,2-trichloroethane, and 1,1,2,2-tetrachloroethane. The average concentrations of toluene, 1,2-dichloropropane, and 1,1,2,2-tetrachloroethane in the post-smoking period were lower than during smoking when the ACH was increased to 2.8 1/h (Fig. 6). The enhanced ventilation would reduce the residential time of the VOCs in the chamber; as a result, less VOCs were re-emitted during the post-smoking period. Even though there was raised concentrations of methylene

chloride and 1,1,2-trichloroethane measured during the post-smoking period, there were clearly a reduction of the absolute increased values for these VOCs. The results have indicated that indoor pollutants could be reduced by enhanced ventilation and thus would minimise formation of secondary VOCs or evaporation of VOCs adsorbed on PM.

CONCLUSIONS

This pilot study has provided a characterisation of the emissions of ETS generated by smoking cigarettes in environmental chamber. Smoking of cigarettes as illustrated

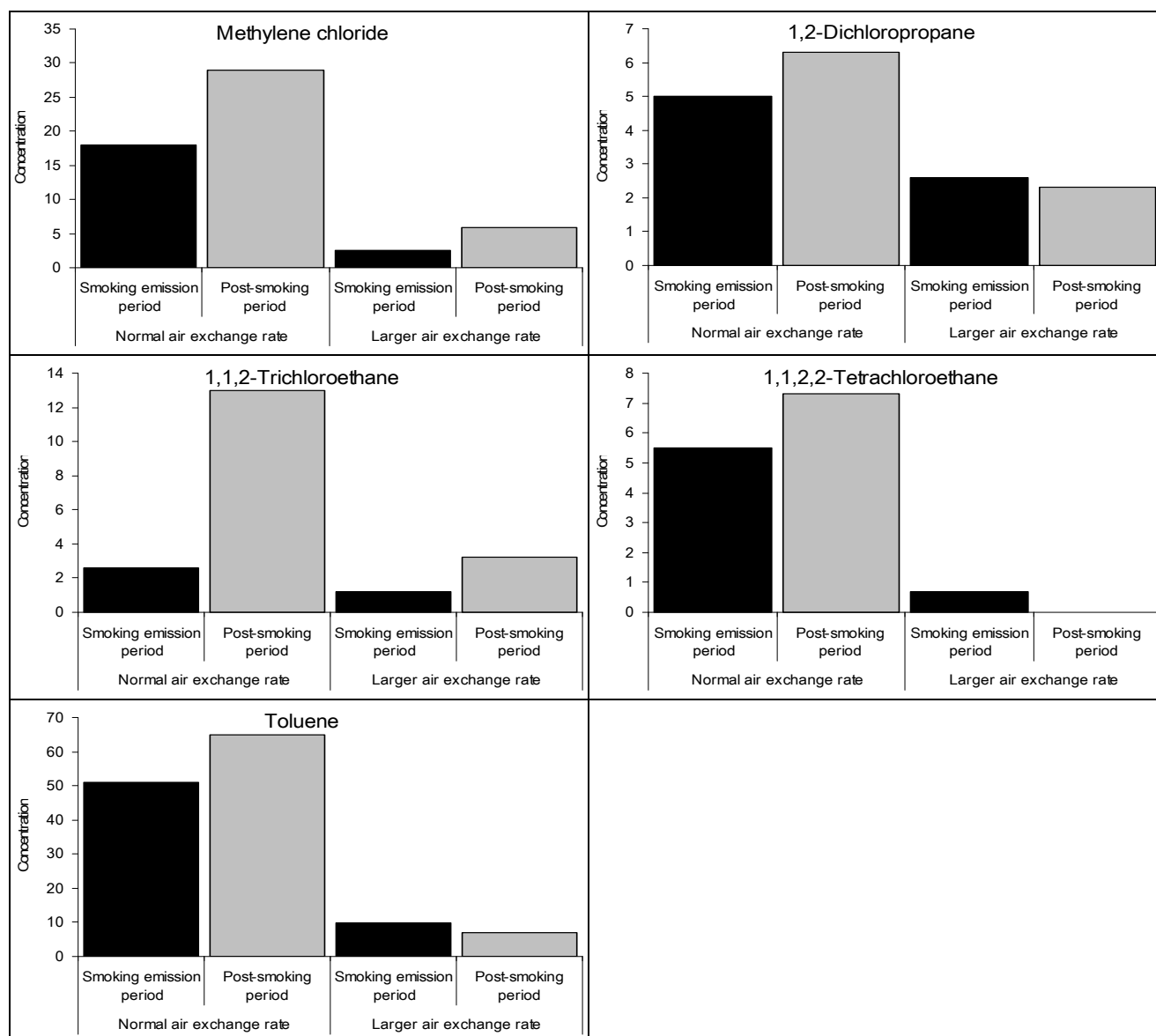


Fig. 6. Comparison of concentrations for the five selective VOCs under normal (0.5 1/h) and larger (2.8 1/h) air exchange flow rates in the environmental chamber study.

by the environmental chamber would generate harmful concentrations of toxic chemicals such as formaldehyde and benzene and would exceed the indoor air quality objectives as set by the IAQ Management Group of the HKEPD. The emission profiles of the ETS generated by smoking could vary due to the different composition of the additives, nicotine, tar, menthol in the content of the cigarettes. The enhanced ventilation could reduce ETS pollutants concentrations and could reduce the resident time of the VOCs, PMs and the other pollutants in the environmental chamber.

Further study could include monitoring on the emissions of other toxic pollutants such as PAHs in both gas- and particulate-phases in ETS generated by cigarettes smoking; and to assess the likely exposure and impact on human health. More in-depth investigation should also be conducted in actual furnished rooms in a real apartment to evaluate the

effects of ETS generated by smoking in a real residential environment in Hong Kong or Mainland of China.

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