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Characterization and health risk assessment of $PM_{2.5}$-bound organics inside and outside of Chinese smoking lounges

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ABSTRACT

$PM_{2.5}$ samples were collected at six indoor public places that contained dedicated smoking lounges. Samples were taken in the smoking lounges, at two indoor locations outside of the lounges, and in outdoor air near the venues. Organic carbon (OC), elemental carbon (EC), and non-polar organic compounds including polycyclic aromatic hydrocarbons (PAHs), *n*-alkanes (*n*-C_{16} to *n*-C_{40}), iso/anteiso-alkanes (C_{29} to C_{33}), hopanes and phthalate esters (PAEs) were quantified. Average $PM_{2.5}$ levels of 170.2±85.9 μg/m^3 in the lounges exceeded limits of 25 μg/m^3 set by World Health Organization (WHO); these levels were 5.4 and 3.9 times higher than those indoors and outdoors, respectively. High ratios of OC to $PM_{2.5}$, OC to EC, and PAHs diagnostic ratios in the lounges indicated contributions from environmental tobacco smoke (ETS). The maximum carbon number (C_{max}) and carbon preference indices (CPI) for *n*-alkanes showed ETS transport from the enclosed lounges to nearby indoor non-smoking areas. Iso/anteiso-alkanes in the lounges were 876.5 ng/m^3, ~80 times higher than outdoor levels. 17α(H)-21β(H),30-norhopane and 17α(H)-21β(H),(22R)-homohopane were much higher in the lounges than outdoor air, but they cannot be directly attributed to ETS. Estimated carcinogenic risks of PAHs in the lounges exceeded the acceptable level of 10^{-6}.

Keywords: Environmental tobacco smoke; Indoor public places; Smoking-free policy; $PM_{2.5}$; PAHs; Iso/anteiso-alkanes

1. Introduction

Environmental tobacco smoke (ETS) from cigarettes, cigars, pipes, and e-cigarettes contains a variety of gases and particles that are detrimental to public health (Kavouras et al., 1998; Bansal and Kim, 2016). Suspended particulate matter (PM), a major component of ETS, contains diverse compounds such as polycyclic aromatic hydrocarbons (PAHs), alkanes, and organonitrates that are genotoxic and carcinogenic (Rogge et al., 1994; Liang and Pankow, 1996). ETS elevates the risk of respiratory diseases and lung cancer for both of children and adults (Kim et al., 2014; Lee et al., 2016). ETS concentrations in entertainment venues are 2.4–18.5 times higher than those in office buildings (Siegel and Skeer, 2003), and increased nicotine metabolites in urine have been found for casino and other hospitality workers (Larsson and Montgomery, 2008; Achutan et al., 2009). Repace (2004) found that ETS generated 50 times more cancer-causing particles than those found along city streets and highways during rush-hour traffic. Acute ETS exposures degrade micro vascular functions (Adamopoulos et al., 2008). General (2010) concluded that exposures to low levels of ETS can increase endothelial dysfunction and inflammation.

Smoke-free policies in the workplace and other indoor public areas intend to reduce the number of smokers and ETS exposures (Bauer et al., 2005; Seo et al., 2011; MacNaughton et al., 2016). After implementation of a smoke-free workplace law (D-2002SFL) in Delaware, USA in 2002, $PM_{2.5}$ mass and $PM_{2.5}$-bound polycyclic aromatic hydrocarbon (PAH) concentrations decreased by factors of 20 to 40, from 205 $\mu g/m^3$ and 163 ng/m^3 to 9 $\mu g/m^3$ and 4 ng/m^3, respectively, in entertainment venues (Repace, 2004). Indoor smoking bans in public buildings (WHO 2015) were introduced in eighteen Chinese megacities in 2014. However, smoking lounges, enclosed public areas such as shopping malls, entertainment venues, and airports, are exempt. Although air in the smoking lounge can be ventilated with enhanced circulation and/or filtration systems, this does not completely eliminate health risks

from ETS exposure.

This study characterizes $PM_{2.5}$ (particles with an aerodynamic diameter less than 10 micrometers [μm]) from ETS inside and outside of six indoor smoking lounges in Hong Kong and Macau, Special Administrative Regions of China during 2016. Chemical profiles for organic carbon (OC), elemental carbon (EC), PAHs, *n*-alkanes, iso/anteiso-Alkanes, phthalate esters (PAES), and hopanes were measured. PAH diagnostic ratios, indices of *n*-alkanes, and pollutant ratios of smoking to non-smoking indoor areas (NSIA) were examined to evaluate similarities, differences, and potential source mixtures. Potential transport from smoking to non-smoking areas is investigated and health risks of PAHs and PAEs are assessed.

2. Methodology

2.1 Site description and sample collection

The six smoking lounges average 192 visitors per day and are used daily, seven days a week. Smoking outside the lounges is forbidden and monitored by security guards. Other pollution within the larger indoor areas derives from outdoor air infiltration, dust raised by foot traffic, and cleaning/maintenance activities. Four sampling locations were selected for each venue, including: a) inside the smoking lounge (SL), b) 2 m from the SL entry/exit ($NSIA_{2m}$), and c) 5 m from the SL entry/exit ($NSIA_{5m}$), and d) outdoor air (~25 m from the venue). Table 1 summarizes the smoking lounge characteristics.

Two collocated $PM_{2.5}$ samples were collected onto quartz-fiber filters (47mm, QMA, Whatman, Clifton, NJ, USA) using mini-volume air samplers (Airmetrics, Eugene, OR, USA) at a flow rate of 5 L min^{-1} for 24 hours (from 08:00 to 07:59 local standard time next day). Four samples were collected at each of the entertainment venues' four sampling locations with a total of 96 samples for the six venues.

Before sampling, filters were pre-fired (780 °C, 3 h) to remove adsorbed organic vapors.

$PM_{2.5}$ mass were obtained by gravimetry using a Sartorius ME 5-Felectronic microbalance (±1μg sensitivity; Sartorius, Göttingen, Germany). Each filter was weighed at least two times before and after sampling which were equilibrated at temperature (22±2 °C) and relative humidity (RH, 35-45 %) controlled room. The maximum differences between the replicates were <15 and <20 μg, for blank and loaded filters, respectively. To prevent any loss of volatiles, samples were packed in pre-baked aluminum foil and stored in a freezer at −20 °C.

2.2 Carbonaceous aerosol analyses

OC and EC were quantified on a 0.53 cm^2 punch from each sample with a DRI model 2001 thermal/optical carbon analyzer (Atmoslytic, Inc., Calabasas, CA, USA) following the IMPROVE_A thermal/optical reflectance protocol (Chow et al., 1993; Ho et al., 2004; Cao et al., 2007; Chow et al., 2007). The IMPROVE_A protocol produces four thermal OC fractions, OC1, OC2, OC3, and OC4 at 140, 280, 480, and 580 °C, respectively, in a pure helium [He] atmosphere. Three thermal EC fractions, EC1, EC2, and EC3 at 580, 740, and 840 °C, respectively, are obtained in a 2% oxygen (O_2)/ 98% He atmosphere. A pyrolyzed carbon fraction, OP, is determined when reflected laser light attains its original intensity after O_2 is added to the carrier gas. OC is defined as the sum of the four OC fractions (OC1-OC4) plus OP, and EC is defined as the sum of the three EC fractions (EC1-EC3) minus OP to account for conversion of OC to EC by pyrolysis.

2.3 Non-polar organic speciation analysis

Non-polar organic compounds were quantified, including PAHs, *n*-alkanes (*n*-C_{14} to *n*-C_{40}), iso/anteiso-alkanes, hopanes and PAEs, using in-injection port-thermal desorption-gas chromatography-mass spectrometry (TD-GC/MS) (Ho and Yu, 2004; Ho et al., 2008). Filter sections (0.53–2.63 cm^2) were inserted into a TD tube for insertion into the GC injector port

at 50°C. The temperature was then raised to 275 °C for desorption in a splitless mode while the GC oven temperature was kept at 30 °C. After the injector temperature reached the set point, the oven program started. The mass spectrometer detector was full-scanned from 50 to 650 amu under electron impact ionization (EI) at a voltage 70 eV and source temperature of 230 °C. Identification was achieved by associating characteristic ion fragments and peak retention times with those of standards. Field blank filters were analyzed using the same procedures.

2.4 Health risk assessment

ETS exposure changes aortic waveforms and weakens microvascular function, even after exposure ends (Argacha et al., 2008). A 12% decrease in heart rate variability from ETS exposures of non-smoking adults to a 53 μg/m^3 increment of $PM_{3.0}$ in a commercial airport was reported by Pope et al. (2001). This was interpreted by Repace et al. (2011) as a 12% increase in cardiac mortality risk, which translates to ~2.3% per 10 μg/m^3 increase in $PM_{3.0}$.

Health risks are estimated from PAH and PAE lifetime average daily doses (LADD) through PM inhalation (Li et al., 2013; Ma et al., 2014; Kong et al., 2015). The incremental lifetime cancer risk (ILCR) is estimated as (Ma et al., 2014; Kong et al., 2015):

$$LADD = C \times IR \times ED \times EF/(BW \times AT) \quad (1)$$

$$ILCR = LADD \times CSF \quad (2)$$

where C is the mass concentration of $PM_{2.5}$ PAHs and PAEs (μg/m^3); IR is the inhalation rate (32.73 m^3/day for adult); ED is the lifetime exposure duration (5 years for workers); EF is the exposure frequency (240 working days/year); BW is the body weight (70 kg for adult); and AT is the average lifetime for carcinogens (70 years × 365 day/year = 25550 days). CSF in Eq. 2 represents the cancer slope factor for bis(2-ethylhexyl)phthalate (DEHP) and benzo[a]pyrene (BaP) inhalation exposure, 0.014 and 3.14 (mg/kg/day)$^{-1}$, respectively. In this

study, the BaP equivalent concentration (BaP_{eq}) was used to calculate LADD instead of mass concentration *C* in Eq. 1. BaP_{eq} was calculated as follows:

$$\sum BaP_{eq} = \sum PAH_i \times TEF_i \quad (3)$$

where BaP_{eq} is the carcinogenic potency of a congener based on BaP-equivalent concentration, TEF is the toxic equivalent factor (Nisbet and Lagoy, 1992). Supplemental Table S1 shows the abbreviations and the TEF values of eighteen PAHs

2.6 Quality assurance and quality control (QA/QC)

Airmetrics mini-volume samplers were checked and calibrated before each sampling event. Field blanks were collected to correct for background levels. Detailed QA/QC procedures for measurements of OC/EC and non-polar organic compounds are documented elsewhere (Ho et al., 2008; Ho et al., 2011; Cao et al., 2013; Xu et al., 2013).

The OC/EC analyzer was calibrated with known quantities of methane gas daily. Replicate analyses were done for each group of ten samples with relative standard deviation (RSD) of <5% for total carbon (TC), and <10% for OC and EC. Average field blanks were 1.72 and 0.09 μg/m^3 for OC and EC, respectively, and subtracted from each sample.

For TD-GC/MS analysis, chrysene-d_{12} ($C_{18}D_{12}$) (98%, Sigma-Aldrich, Bellefonte, PA, USA) was added as an internal standard (IS) for the PAH and PAEs, and *n*-tetracosane-d_{50} (*n*-$C_{24}D_{50}$) (98%, Aldrich, Milwaukee, WI, USA) was used for the *n*-alkanes, iso/anteiso-alkanes and hopanes. A six-point calibration over a concentration range of 0.5–10.0 ng for each of the target compounds was determined from standard mixtures (Sigma-Aldrich, Bellefonte, PA, USA), and the coefficients of variation (R^2) for linear regressions of the calibration curves were >0.99.

3. Results and Discussion

3.1 $PM_{2.5}$ mass concentrations

As shown in Table 2, the highest $PM_{2.5}$ levels were found in the lounges, ranging from 65.7 - 297 μg/m^3 with an average of 170.2±85.9 μg/m^3. These averages were 5.4 and 3.9 times, higher than those at NSIA and outdoor environments, respectively. These levels exceeded the 25-75 μg/m^3 guideline recommended by international environmental departments and health agencies for indoor air quality (JGJ/T309-2013; ANSI, 2007; WHO, 2006). Outdoor $PM_{2.5}$ levels were also high, ranging from concentrations 31.1–65.1 μg/m^3 with an average of 44.2±14 μg/m^3, often exceeding U.S. 24-h national ambient air quality standards of 35 μg/m^3.

These concentrations are consistent with previous studies shown in Table S2. $PM_{2.5}$ concentrations in the non-smoking areas (i.e., $NSIA_{2m}$ and $NSIA_{5m}$), ranging from 28.8-32.9 μg /m^3, were higher than those of measured in U.S. non-smoking indoor public areas of 3.1-22.3 μg/m^3 (Jiang et al., 2011; Repace et al., 2011). With respect to the smoking lounges, this study also showed $PM_{2.5}$ levels 1.8 times higher than Nafees et al. (2012) for Pakistan and 2.7-3.2 times higher than those reported by Repace et al. (2011) and Jiang et al. (2011) for the United States.

3.2 OC and EC

Table 3 shows average OC levels in the lounges were 133.1.1±69.0 μg/m^3, accounting for 94.1% of TC, whereas TC accounts for over 83% of $PM_{2.5}$. TC levels in the lounges were 7.8-9.7 times those at indoor and outdoor locations. The average OC/EC ratio was 15.8±3.9 in the lounges, decreasing with distance outside of the lounges: 5.19±1.7 at $NSIA_{2m}$ and 4.3±0.7 at $NSIA_{5m}$.

Abundances of the thermal fractions differ by source type (Watson et al., 1994; Chow et al., 2004). Figure 1 shows that OP in SLs accounted for 17.9% of TC, much higher than those found outdoors, consistent with smoldering emissions from biomass burning (Andreae and Gelencser, 2006). OC1 is an indicator of semi-volatile organic compounds (SVOCs) that may evaporate with aging as the gas fraction decreases with dilution. Higher EC1 and EC2 found outdoors are indicators of engine exhaust (Watson et al., 1994). Table S3 shows strong correlations ($r > 0.9$ with $p < 0.05$) between $PM_{2.5}$ mass and most organic carbon fractions (i.e., OC1 to OC3, plus OP), but a negative correlation with OC4 ($r = -0.77$, $p = 0.10$).

3.3 PAHs

As shown in Figure 2, total quantified PAH concentrations (∑TPAHs) in the lounges (217.42±63.05 ng/m^3) were ~6-8 times those at $NSIA_{2m}$ and $NSIA_{5m}$ (28.07±9.67, 36.53±19.22 ng/m^3, respectively) and ~5 times higher than outdoors (46.53±33.41 ng/m^3). The SL PAHs were 1.4 times those found in a U.S. casino prior to a smoking ban (Repace et al., 2011). Total PAHs at the NSIA locations (14.52-62.58 ng/m^3) were also ten times those found at the U.S. casino after the smoking ban.

The three most abundant PAHs inside the lounges are phenanthrene (PHE), chrysene (CHR), and benzo[a]anthracene (BaA), with average concentrations of 39.3±35.6, 40.0±27.6, 23.5±11.6 ng/m^3, respectively, accounting for 18.1, 18.4, 10.8 % of ∑TPAHs. On average, the sum of middle molecular weight PAHs ($\sum PAHs_{MMW}$, with 4 aromatic rings) was higher than that of low molecular weight PAHs ($\sum PAHs_{LMW}$, with 2-3 aromatic rings) and high molecular weight PAHs ($\sum PAHs_{HMW}$, with 5-6 aromatic rings) inside the lounges, indicating the contrast between smoking and nonsmoking environments (Figure 3).

Average benzo[a]pyrene (BaP) was 10.1 ng/m^3 in the SLs, accounting for 4.9% of the ∑PAHs and 5.4~10.2 times higher than BaP at the NSIA locations and 10 times the World

Health Organization's (WHO) indoor air quality guideline of 1.0 ng/m^3.

PAHs diagnostic ratios have been used to identify potential pollution sources (Table 4) (Yunker et al., 2002). The ratios of anthracene (ANT) to (ANT + PHE) >0.10 and BaA/(BaA + CHR) > 0.35 suggest combustion source contributions at the indoor and outdoor locations (Tobiszewski and Namieśnik, 2012). Owing to the different PAH volatilities, these ratios may change with aging, which introduces uncertainty for quantitative source attribution (Ding et al., 2012). Indoor fluoranthene (FLU) to perylene (PYR) ratios (i.e., FLU/[FLU+PYR]) exceeded 0.5 inside and outside of the lounges, which has been attributed to biomass burning (Katsoyiannis et al., 2011). The largest difference is the enhancement of the BaP to benzo[ghi]perylene (BghiP) ratio in the lounges compared to those measured outside, indicating that this is a good ETS marker to estimate contribution of tobacco combustion (Zhang et al., 2009).

3.4 *n*-alkanes and iso/ anteiso-alkanes

C_{16} to C_{40} *n*-alkane homologue levels are illustrated in Figure 2. The highest total *n*-alkanes ($\sum$*n*-alkanes) (2554.3 ng/m^3) in the lounges were ten times higher than at the other sampling points. Different profiles were observed between indoor and outdoor samples. The maximum carbon number (C_{max}) of C_{31} was found for the lounges and NSIA samples, compared with outdoor C_{max} of C_{24}. The heavier *n*-alkanes (i.e., *n*-C_{24} to *n*-C_{33}) accounted for 89.3% and 69.8% of $\sum$*n*-alkanes in the lounges and outdoors, respectively.

The Carbon Preference Indices (CPI) for *n*-alkanes is:

$$\mathrm{CPI} = \sum C_{17} - C_{39} / \sum C_{16-}C_{40} \qquad (4)$$

CPI≈1 indicates contributions from vehicular emissions and other anthropogenic activities, while CPI>1 for epicuticular waxes from of terrestrial plants (Bray and Evans, 1961; Choi et al., 2015; Wang et al., 2016). The CPI=2.97 in the lounges exceeded the NSIA CPIs

(1.28~1.31) and was much higher than the outdoor CPI (1.02) (Table 4). C_{max} and CPI values are clear indicators of the tobacco biomass burning at the indoor sites in contrast to the typical fossil fuel burning values measured outside (Rogge et al., 1993, 1994).

Due to their low reactivity, iso/anteiso-alkanes have long atmospheric residence times and are stable markers of ETS contributions. Kavouras et al. (1998) used ratios of the most abundant *n*-alkanes and iso/anteiso-alkanes to identify ETS contributions in ambient air. Indoor iso/anteiso-alkanes with carbon numbers of C_{29} to C_{34} show a characteristic profile (Figure 2) that is similar to that of tobacco leaf surface wax (Schauer et al., 2007). The average total iso/anteiso-alkanes (∑iso/anteiso-alkanes) level in the lounges was 1392.8±505.2 ng /m^3, two orders of magnitudes higher than the outdoor average (11.8±10.4 ng /m^3) (Figure 2). ∑iso/anteiso-alkanes at the NSIA locations were 29.4-32.4 times higher than the outdoor values, indicative of ETS penetrating from the lounges into the indoor non-smoking areas. Iso-C_{31} and anteiso-C_{32} were the most abundant species from ETS in iso/anteiso-alkanes. As shown in Table 5, large differences in diagnostic ratios of iso/anteiso to straight chain *n*-alkanes were found between indoor and outdoor samples (Table 5). Higher alkane ratios indoors than outdoors further support the effect of ETS, consistent with findings of Bi et al. (2005).

3.5 PAES

Six PAEs (i.e., dimethylphthalate, [DMP]; diethyl phthalate, [DEP]; di-n-butyl phthalate, [DBP]; bis(2-ethylhexyl)phthalate, [DEHP]; di-n-octyl phthalate, [DNOP]; and one plasticizer of di-2-ethylhexyladipate [DEHA]) were quantified (Figure 2). Total PAEs (∑PAEs) concentrations were 926.5 - 1391.2 ng/m^3 indoor and 610.4 ng/m^3 outdoor. As shown in Table S4, ∑PAEs were negatively correlated with ∑iso/anteiso-alkanes (r= -0.54) and OC (r= -0.6), consistent with ETS not being a major source. PAEs are used in personal care products,

plastics (including polyvinylchloride [PVC]), floorings, and wall coverings (Afshari et al., 2004; Ejaredar et al., 2015; Myridakis et al., 2015). DEHP was the dominant species measured indoors and outdoors, accounting for 33-67% of the ∑PAEs, followed by DBP (21-45%). DEHP is the most common PAE in PVC, while DBP is a component of latex adhesives (Mariana et al., 2016).

3.6 Hopanes

Total hopane (ΣHopanes) levels of 11.25 ng /m^3 in the lounges were 7.9 times higher than outdoors (1.43 ng/m^3) and 12.8 times higher than those found in non-smoking indoor areas (0.17 to 1.09 ng /m^3). Hopanes are common in petroleum products and are most often used as markers for lubricating oils in engine exhaust emissions. Two hopanes, 17α (H)-21β(H),30-norhopane and 17α(H)-21β(H),(22R)-homohopane, were most highly enriched over the outdoor levels. There is no other evidence of these hopanes originating in biogenic combustion, and more specific measurements of confined ETS would be needed to classify these as potential markers for this source.

3.7 Comparison of smoking and non-smoking concentrations

Ratios of smoking lounge to indoor locations (i.e., $SL/NSIA_{2m}$ and $SL/NSIA_{5m}$) are shown in Figure 4. Most ratios were much greater than unity with ratios of PAEs, DMP, DEHP, DNOP and DEHA near unity, indicating a uniform source mixture for these compounds. Owing to different levels of PAH volatility, SL/NSIA ratios for low molecular weight PAHs (i.e., PAH_{LMW}) were slightly lower than those for medium- and high-molecular weight PAHs. Higher than unity ratios were also found for iso/anteiso-alkanes and hopanes (i.e., 17α(H)-21β(H),30-norhopane and 17α(H)-21β(H),(22R)–homohopane).

3.8 Health risk assessment

Difference in $PM_{2.5}$ concentrations between the smoking lounge (170.2 μg/m^3) and outdoors (44.2 μg/m^3) was 126.0 μg/m^3. Using the acute health risk of 2.3% per 10 μg/m^3 increase in $PM_{3.0}$, the estimated acute health risks for 1-2 hour ETS exposure would be 29% (i.e., 126.0 μg/m^3 x 2.3%/10 μg/m^3). About half the samples acquired in the lounges were at or above this level. The high percentage proves that ETS poses an acute threat to the health of smokers and workers in lounges, even with short exposure durations (<1 h).

3.8.1 Cancer risk of PAHs

The PAH total carcinogenic potency, BaP_{eq}, for smoking lounge inhalation exposure was 20.5±8.0 ng/m^3, while lower values were found at NSIA locations, ranging from 2.2±1.7 to 3.4±2.6 ng/m^3, close to the outdoor levels of 3.6±3.9 ng/m^3. The incremental lifetime cancer risk (ILCR) was 1.4×10^{-6}, which is 6-10 times the risk at NSIA exposure levels (1.5×10^{-7} to 2.3×10^{-7}) and 5.6 times outdoors (2.5×10^{-7}). Smoking lounge risks exceed the acceptable level of 10^{-6}.

Risks are most potent for dibenz[a,h]anthracene (DahA), followed by BaP and a group of tobacco combustion products (e.g., BaA, benzo[b]fluoranthene [BbF], benzo[k]fluoranthene [BkF], and PER). Considering the high TEF values, DahA and BaP contributed to 57%~65% of the total carcinogenicity in $PM_{2.5}$. Such contributions imply that the potential carcinogenicity of $PAHs_{HMW}$ is higher than that of $PAHs_{LMW}$ for biomass combustion, consistent with previous studies (Shen et al., 2013; Jarvis et al., 2014; Hanahati et al., 2015; Mohammed et al., 2016). Even though non-smokers near the lounges experienced lower exposures and their ILCRs were below thresholds, they were still higher than they would have been in a non-smoking environment.

3.8.2 Risk assessment of PAEs

LADD estimates of DEHP in a descending order of sampling locations were: SL (1.4×10^{-6} mg/kg/d) > NSIA (1.2×10^{-6} to 1.3×10^{-6} mg/kg/d) > outdoor (4.7×10^{-7} mg/kg/d). The carcinogenic risks of DEHP in the lounges was 1.9×10^{-8}, close to 1.6×10^{-8} to 1.9×10^{-8} at NSIA locations and higher than the outdoor value of 6×10^{-9}. The estimated carcinogenic risks of inhalation of DEHP were below the acceptable level of 10^{-6}.

4. Conclusion

ETS leads to excessive levels of $PM_{2.5}$ and organic toxics at indoor public places. Although smoking lounges limit the major exposure to those who chose to enter and smoke in these facilities, ETS is not completely confined to these spaces. Pollution generated within the lounges escapes to indoor public areas where non-smokers are exposed.

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488

489 **Table 1. Summary of smoking lounge* characteristics**

	Indoor Public Places					
	I	II	III	IV	V	VI
Smoking lounge area (m^2)	6	50	23	20	12	50
No. of people accessing per hour	5	1	10	12	6	16
Door openings per hour	10	5	20	20	10	30

490 *The smoking lounges sampled are not identified due to a privacy agreement.

491

492 **Table 2. $PM_{2.5}$ concentrations ($\mu g/m^3$) at different locations for each venue**

	Indoor Public Places						
Sampling point	I	II	III	IV	V	VI	Average±Standard Deviation
Smoking Lounge (SL)	120.9	65.7	118.3	238.7	180.9	297.0	170.2±85.9
$NSIA_{2m}$[a]	23.7	31.3	32.2	42.7	33.4	34.2	32.9±6.1
$NSIA_{5m}$[a]	15.8	28.7	36.2	30.3	26.9	34.6	28.8±7.2
Outdoor	44.8	65.1	48.2	31.8	31.1	43.6	44.2±14.0

493 [a]$NSIA_{2m}$ and $NSIA_{5m}$ are non-smoking indoor areas 2m and 5m from the smoking lounge entry/exit.

494 **Table 3. Average $PM_{2.5}$ TC, OC, and EC concentrations.**

	Smoking lounge (SL)	$NSIA_{2m}$	$NSIA_{5m}$	Outdoor
TC(μg /m^3)	141.5±71.8	20.2±3.9	17.8±2.2	18.7±5.5
OC(μg /m^3)	133.1±69.0	17.0±3.3	14.4±2.0	13.7±5.9
EC(μg /m^3)	8.4±3.1	3.3±1.2	3.4±0.5	5.0±1.7
OC/EC	15.8±3.9	5.19±1.7	4.3±0.7	2.7±2.3

495

496 **Table 4. PAHs diagnostic ratios, indices of *n*-alkanes, and cancer risks at indoor and outdoor**
497 **locations.**

Diagnostic ratios or Indices *	Smoking Lounge (SL)	$NSIA_{2m}$	$NSIA_{5m}$	Outdoor
ANT/(ANT + PHE)	0.14	0.18	0.12	0.13
BaA/(BaA + CHR)	0.37	0.42	0.44	0.54
Flu/(Flu + PYR)	0.51	0.55	0.55	0.51
BaP/BghiP	1.18	0.47	0.39	0.64
C_{max}	C31	C31	C31	C24
CPI	2.97	1.31	1.28	1.02

498 * Abbreviations for PAHs and indices for *n*-alkanes are specified in the text.

499

500 **Table 5. Comparison of diagnostic ratios for iso/anteiso-alkanes and *n*-alkanes.**

	This study				(Bi et al., 2005)
Alkanes*	Smoking lounge (SL)	$NSIA_{2m}$	$NSIA_{5m}$	Outdoor	Office
a-C30/n-C30	2.1	0.9	1.2	0.3	2.51± 0.53
i-C31/n-C31	0.5	0.4	0.5	0.2	0.53 ± 0.04
a-C32/n-C32	2.0	1.6	2.0	0.6	3.11 ± 0.44
i-C33/n-C33	0.5	0.4	0.5	0.3	0.44 ± 0.05
a-C30/i-C31	0.7	0.8	0.8	0.8	0.68 ± 0.08
i-C33/i-C31	0.5	0.5	0.4	0.5	0.25± 0.07
a-C32/i-C33	1.8	2.6	2.8	2.2	3.44± 0.58
$\sum$ (i+a)-Cn/$\sum$n-Cn	0.9	0.7	0.9	0.4	0.94 ± 0.10

501 * i represents iso-, a- represents anteiso-, and n- represents straight chain of alkane.

502

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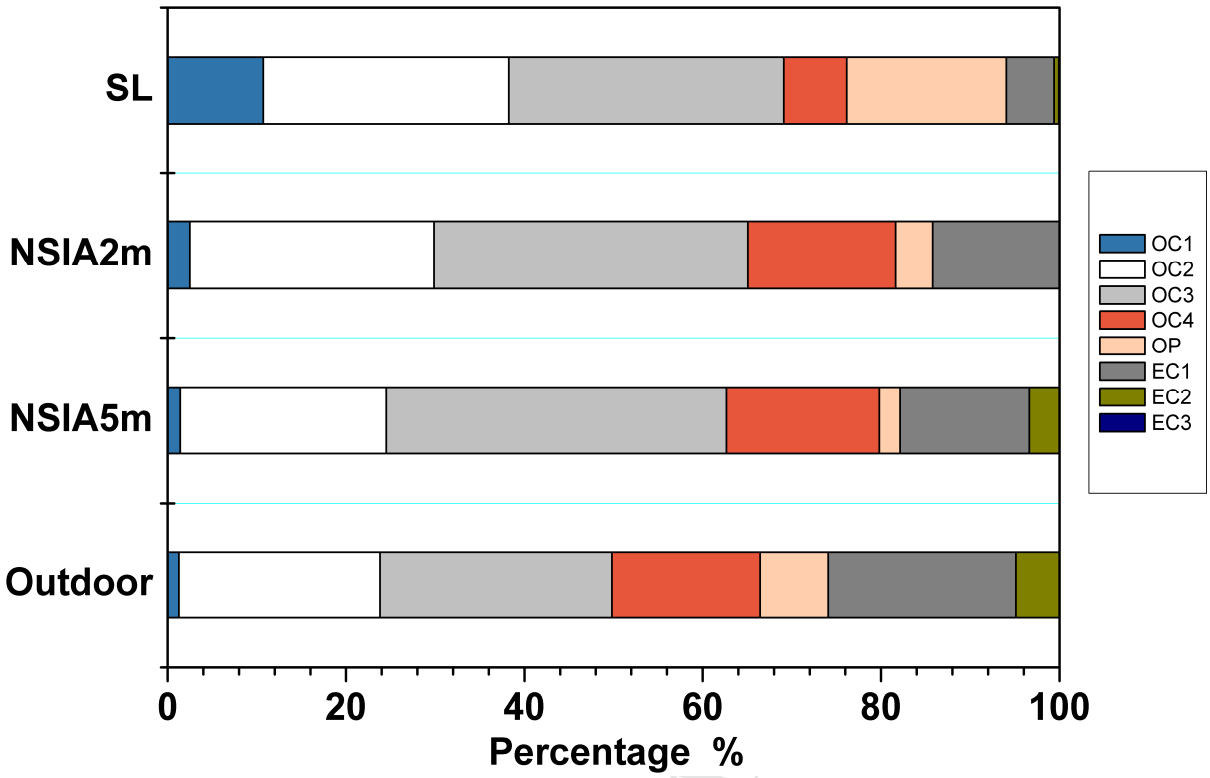


504

505 **Figure 1. $PM_{2.5}$ carbon fraction abundances and fraction of total carbon.**

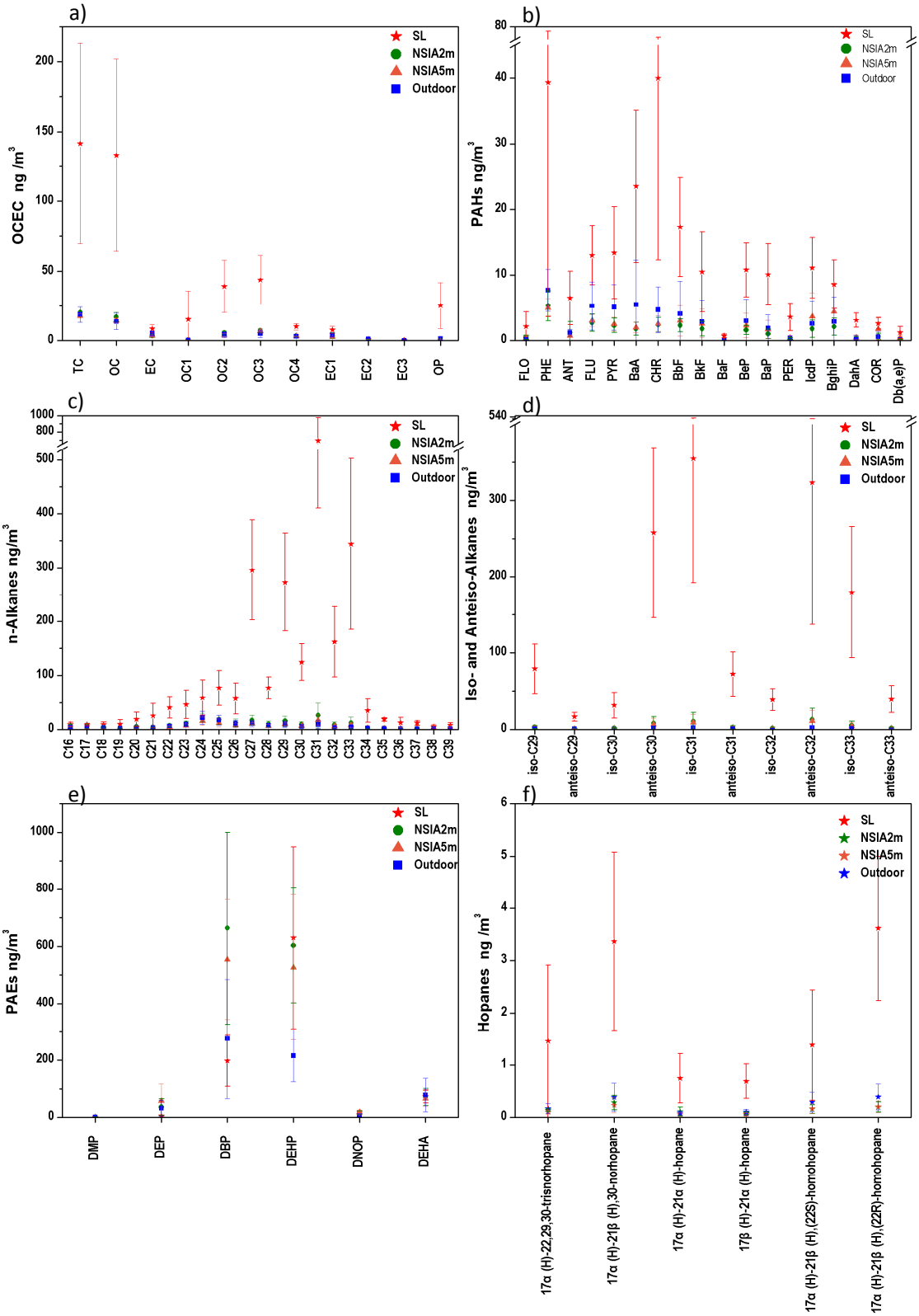
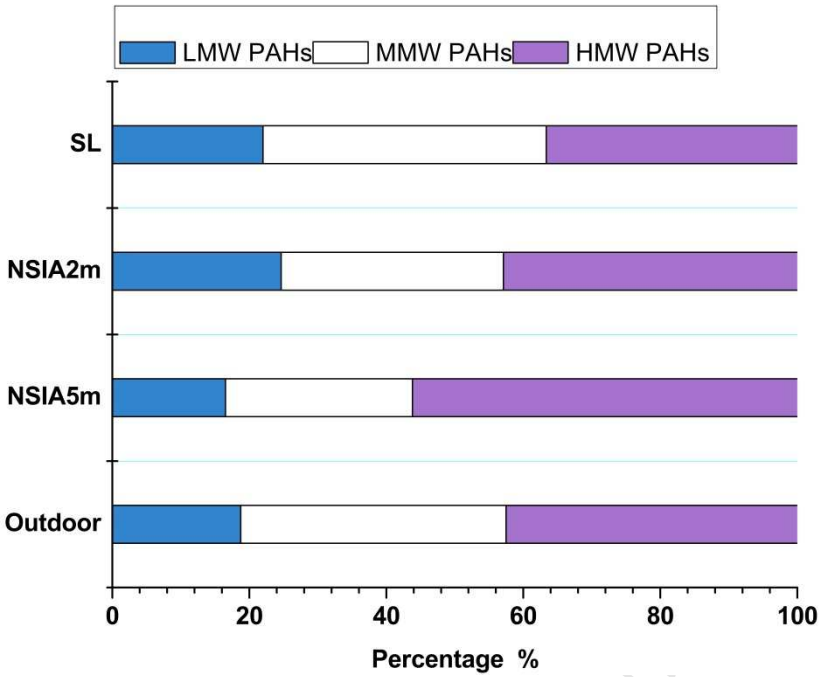


Figure 2. Average and ranges of: a) OC and EC, b) PAH, c) *n*-alkanes, d) iso- and anteiso-alkane, e) PAE, and f) hopane concentrations in $PM_{2.5}$ inside and outside of the smoking lounges. PAH and PAE abbreviations are listed in Table S1 and the text.



510

511 **Figure 3. Fractions of low, medium, and high molecular weight PAHs in total quantified PAHs.**
512 **LMW-PAHs (with 2-3 aromatic rings) is the sum of FLO, PHE, and ANT; MMW-PAHs (with 4 aromatic**
513 **rings) is the sum of FLU, PYR, BaA, and CHR; HMW-PAHs (with 5-6 aromatic rings) is sum of BbF, BkF,**
514 **BaF, BeP, BaP, PER, IcdP, BghiP, DahA, COR, and DaeP. See Table S1 for compound names.**

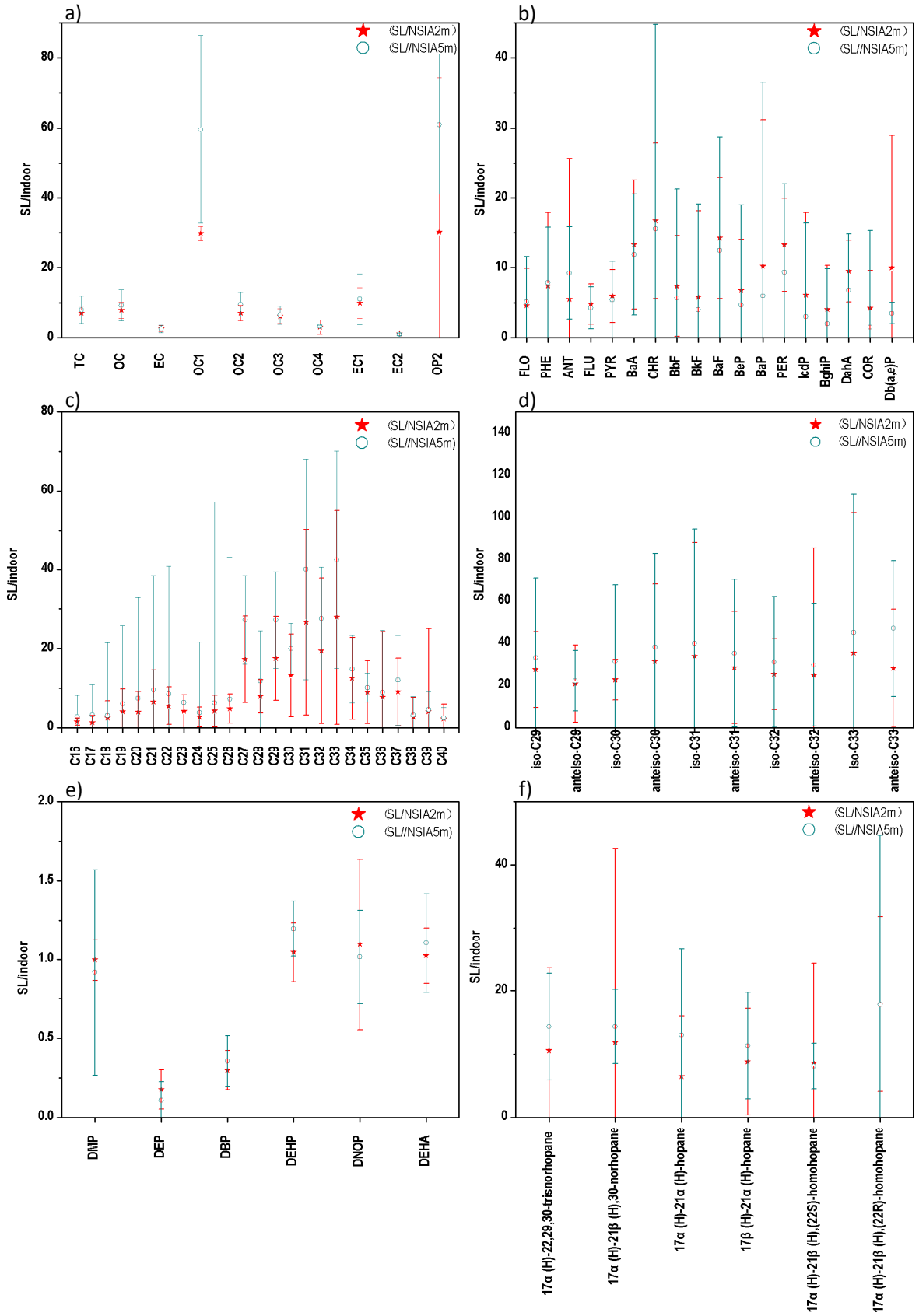


Figure 4. Ratios of smoking lounge to indoor locations (i.e., $NISA_{2m}$ and $NISA_{5m}$) for: a) OC and EC, b) PAH, c) *n*-alkanes, d) iso- and anteiso-alkane, e) PAE, and f) hopane concentrations in $PM_{2.5}$ inside and outside the smoking lounges. PAHs and PAEs abbreviations are listed in the Table S1 and the text.

519 **Table S1. Abbreviation definitions and Toxic Equivalent Factors (TEF) for the eighteen**
520 **polycyclic aromatic hydrocarbons (PAHs).**

Compounds	Abbreviation	Toxic Equivalency Factor (TEF)
fluorene	FLO	0.001
phenanthrene	PHE	0.001
anthracene	ANT	0.01
fluoranthene	FLU	0.001
pyrene	PYR	0.001
benzo[a]anthracene	BaA	0.1
chrysene	CHR	0.01
benzo[b]fluoranthene	BbF	0.1
benzo[k]fluoranthene	BkF	0.1
benzo[a]fluoranthene	BaF	-
benzo[e]pyrene	BeP	-
benzo[a]pyrene	BaP	1
perylene	PER	0.1
indeno[1,2,3-cd]pyrene	IcdP	0.1
benzo[ghi]perylene	BghiP	0.01
dibenzo[a,h]anthracene	DahA	1
coronene	COR	-
dibenzo(a,e)pyrene	DaeP	-

521

522 **Table S2. Comparison of $PM_{2.5}$ concentrations (μg/m^3) found in different entertainment venues**
523 **(EVs).**

Venue	Venue Location	Indoor Smoking Area	Indoor Non-smoking Area	Outdoor Environment
Gaming zones (Nafees et al., 2012)	Pakistan	93±45.3	-	30±8.5
Smoking EVs (Repace et al., 2011)	United States	53.8	3.1	4.3
Smoking EVs (Jiang et al., 2011)	United States	63.2	22.3	6.7
This study	Hong Kong and Macau	170.2±85.9	32.9±6.1/28.8±7.2	44.1±14.0

524
525

526 **Table S3. Spearman correlation coefficients between $PM_{2.5}$ mass and organic/elemental carbon**
527 **(OC/EC) fractions in smoking lounges**

	OC1	OC2	OC3	OC4	OP	EC1	EC2	EC3
r	1.00	1.00	0.94	-0.77	0.94	0.83	0.77	0.88
p	0.003	0.003	0.02	0.10	0.02	0.06	0.10	0.67

528 **Table S4. Spearman correlation coefficients of ∑iso/anteiso-alkanes and OC with ∑PAEs in**
529 **smoking lounges**

	∑iso/anteiso-alkanes	OC
r	-0.54	-0.60
p	0.27	0.21

530
531

1. Although confining smokers to an enclosed area reduces exposure to non-smokers, environmental tobacco smoke (ETS) still migrates to indoor air for non-smokers.
2. The ratio of benzo[a]pyrene to benzo[ghi]perylene is much higher in the smoking area and is a useful diagnostic ratio for determining the presence of ETS.
3. Despite exposure to ETS, the health risk to non-smokers outside of the smoking lounges did not exceed guidelines for the areas studied.