https://doi.org/10.1016/j.scitotenv.2020.139684

Effects of Indoor Activities and Outdoor Penetration on $PM_{2.5}$ and Associated Organic/Elemental Carbon at Residential Homes in Four Chinese Cities during Winter

Zhuo-Zhi Zhang[1], Yuan Gao[1], Qi Yuan[1], Yan Tan[1], Hai-Wei Li[2], Long Cui[3], Yu Huang[3], Yan Cheng[4], Guang-Li Xiu[5], Sen-Chao Lai[6], Judith C. Chow[7], John G. Watson[7], Shun-Cheng Lee[1*]

[1] Department of Civil and Environmental Engineering, The Hong Kong Polytechnic University, Hung Hom, Hong Kong, China

[2] Jiangsu Key Laboratory of Atmospheric Environment Monitoring and Pollution Control (AEMPC), Collaborative Innovation Centre of Atmospheric Environment and Equipment Technology (CIC-AEET), School of Environmental Science and Engineering, Nanjing University of Information Science & Technology, Nanjing, 210044, China

[3] Key Lab of Aerosol Chemistry & Physics, SKLLQG, Institute of Earth Environment, Chinese Academy of Sciences, Xi'an, 710049, China

[4] Department of Environmental Science and Technology, School of Human Settlements and Civil Engineering, Xi'an Jiaotong University, Xi'an, 710049, China

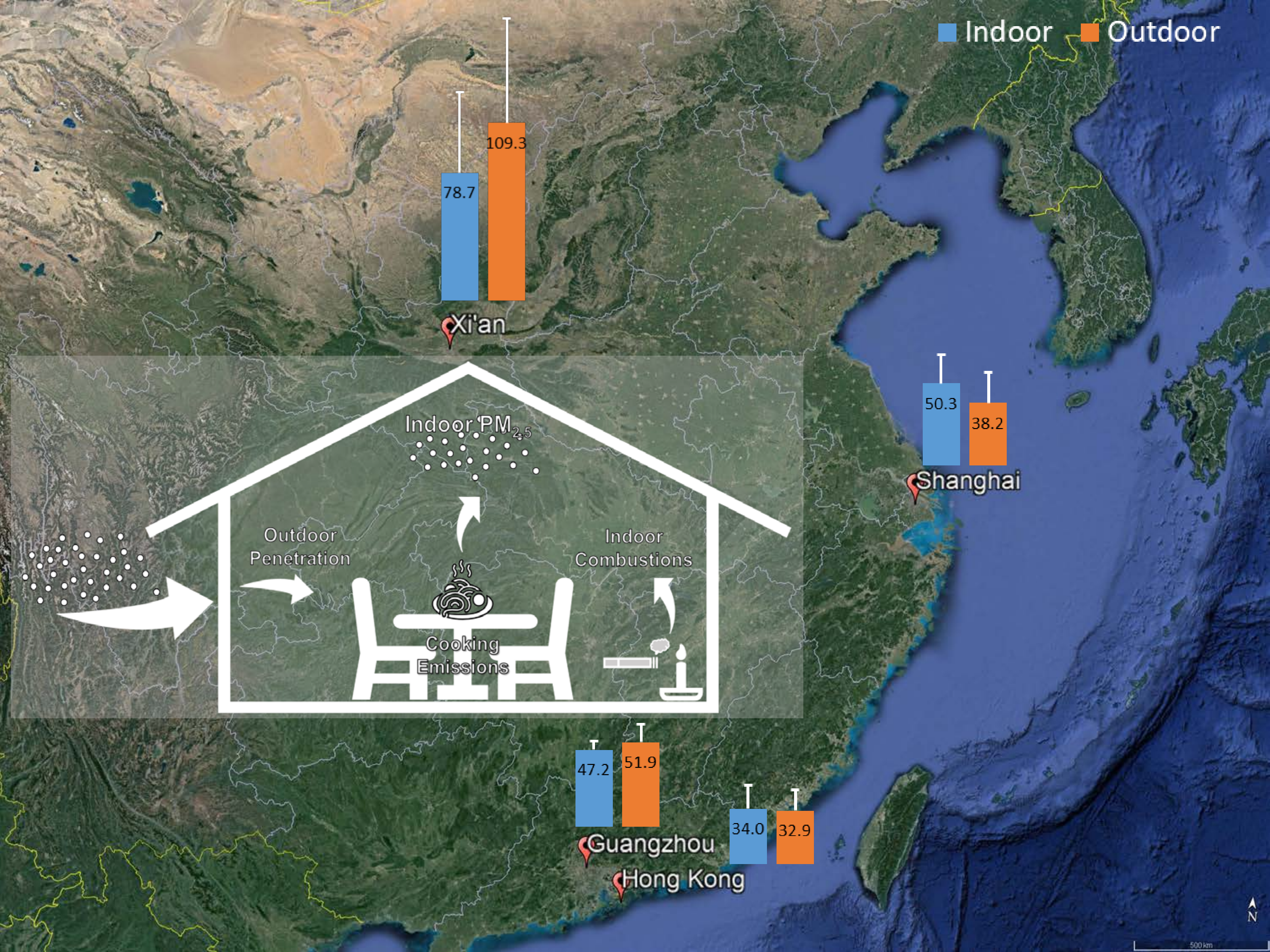
[5] School of Resources and Environmental Engineering, East China University of Science and Technology, Shanghai, 200237, China

[6] School of Environment and Energy, South China University of Technology, Guangzhou, 510006, China

[7] Division of Atmospheric Sciences, Desert Research Institute, Reno, NV 89512, USA

[*] Corresponding author. Tel: 852-2766-6011; Fax: 852-2334-6389

E-mail address: shun-cheng.lee@polyu.edu.hk



Indoor
Outdoor
78.7
109.3
Xi'an
50.3
38.2
Shanghai
47.2
51.9
Guangzhou
34.0
32.9
Hong Kong
Indoor PM2.5
Outdoor Penetration
Indoor Combustions
Cooking Emissions
N
500 km

Highlights

- A general trend of Xi’an>Shanghai>Guangzhou>Hong Kong of indoor $PM_{2.5}$ was found.
- OM accounts significant portion of indoor $PM_{2.5}$, with presence of SOC.
- Indoor cooking was found inducing an increment of $PM_{2.5}$ level of 13.2 μg m^{-3}.
- Tobacco smoking/incense burning could lead to increased $PM_{2.5}$ level of 11.8 μg m^{-3}.
- Impact of outdoor penetration could still be significant with limited air exchange.

Abstract

Increasing public attention on exposure to $PM_{2.5}$ are essential to the assessment of the related health effects. The indoor $PM_{2.5}$ mass and organic/elemental carbon (OC/EC) during winter of 2016-2017 at 68 residential households in four large Chinese cities (i.e. Hong Kong, Guangzhou, Shanghai, and Xi'an) were studied. Average indoor $PM_{2.5}$ varied by two-fold, lowest in Hong Kong (34.0 ± 14.6 μg m^{-3}) and highest in Xi'an (78.7 ± 49.3 μg m^{-3}), with comparable levels for Guangzhou (47.2 ± 5.4 μg m^{-3}) and Shanghai (50.3 ± 17.9 μg m^{-3}). Lowest air exchange rate (AER, 0.8 ± 0.8 h^{-1}) and $PM_{2.5}$ indoor/outdoor (I/O) ratio (0.72 ± 0.23) were found for Xi'an households, indicating the limited influence from indoor sources, while importance of indoor $PM_{2.5}$ sources is signified with the highest $PM_{2.5}$ I/O ratio (1.32 ± 0.43) identified for Shanghai households. For households in four cities, OC and EC accounted for 29.5 % – 38.5 % and 7.5 % – 8.9 % of the indoor $PM_{2.5}$ mass, indicating the significance of carbonaceous aerosols. Larger differences between indoor and outdoor OC (2.6 – 8.4 %) than EC (-2.2 – 1.5 %) indicate the presence of indoor OC sources. Decreasing trends of $PM_{2.5}$ I/O ratio and indoor OC proportion were found as the worsening ambient air quality. On average, 11.8 μg m^{-3} (23.1 %) and 3.02 μg m^{-3} (18.7 %) higher indoor $PM_{2.5}$ and OC concentrations were identified for households with other indoor combustions (e.g., tobacco smoking, incense burning) compared to those with only cooking activities. For Hong Kong and Shanghai households, increments of 13.2 μg m^{-3} (54.1 %) of $PM_{2.5}$ and 4.1 μg m^{-3} (45.4 %) of OC were found at households with cooking activities as compared to households with no specific indoor combustion.

Keywords: Residence; $PM_{2.5}$ (Fine Suspended Particulate); Carbonaceous Aerosols; Indoor Combustion; Ambient Penetration.

1 Introduction

Public attention on air quality is on a rising trend, due to the increased occurrence of severe air pollution events and associated adverse health impacts (Shen et al., 2017; Tian et al., 2016). During a severe haze episode over northern China in 2013, particulate matter (PM) with aerodynamic diameter less than 2.5 μm ($PM_{2.5}$) level was about 9-fold higher than the Chinese National Ambient Air Quality Standard (Huang et al., 2014). These elevated $PM_{2.5}$ concentrations were related to increased hospital visits due to cardiovascular and respiratory illness (Chen et al., 2013). Because of the small size, $PM_{2.5}$ can penetrate into human respiratory system and interfere the lung function (Li et al., 2017; Penttinen et al., 2001a; Penttinen et al., 2001b). Epidemiological studies demonstrated that PM-exposure is associated with numerous respiratory system related diseases, and even with mutagenic and carcinogenic health effects (Achilleos et al., 2017; Heo et al., 2014; Li et al., 2017; Pope III et al., 2002; Pope III and Dockery, 2006). Measurements from the central monitoring site may not be adequate to evaluate PM-induced health risk, as people spend 90% of time indoor, and among which 80% in residences (Klepeis et al., 2001). Indoor $PM_{2.5}$ can be attributed to penetration from ambient through ventilation and infiltration (Cheng et al., 2017; Morawska et al., 2001), as well as various indoor sources including cooking, tobacco smoking, incense burning, cleaning and etc. (Eatough et al., 1989; He et al., 2004; Lee and Wang, 2004; Li et al., 2017; Liu et al., 2016; Moriske et al., 1996; Secrest et al., 2017). This results in higher indoor than ambient $PM_{2.5}$ concentrations (Barraza et al., 2014; Cao et al., 2005; Custódio et al., 2014; Lai et al., 2010; Patterson and Eatough, 2000). It is essential to examine the indoor PM levels in different microenvironments, for further evaluation of PM health impacts.

Carbonaceous aerosol, including organic carbon (OC) and elemental carbon (EC), accounts for 20 to 50% of $PM_{2.5}$ mass (Abt et al., 2000; Cao et al., 2013; Cao et al., 2007; Cao et al., 2003;

Funasaka et al., 2000; Ho et al., 2002; Nunes and Pio, 1993; Zhou et al., 2012). During winter, higher frequency of severe PM pollution events were attributed to carbonaceous aerosols (Wang et al., 2012; Xu et al., 2015; Zhou et al., 2014). OC includes polycyclic aromatic hydrocarbon (PAH) and other organic compounds, some of these components has been classified as human carcinogen (such as Benzo[a]pyrene) by International Agency for Research on Cancer (IARC). EC has also been associated with the increment of mortality related to lung cancer and other respiratory diseases (Frazer, 2002). As cooking and other combustion activities could contribute to the elevated indoor carbonaceous aerosol level, it is important to identify the dominated emission sources to improve indoor air quality.

Levels of impacts on indoor air quality may vary due to the spatial variations of ambient air pollutants concentrations (Huang et al., 2014; Zhang et al., 2012; Zhang and Cao, 2015). Moreover, various climate, heating/ventilation, and cooking activities in different regions may also affect the indoor air quality (Abdullahi et al., 2013; He et al., 2004). Therefore, the characteristics and contributions of indoor emission sources to $PM_{2.5}$ mass and carbonaceous species may exhibit spatial variations, that needs to be further investigated (Cao et al., 2005; Ho et al., 2004; Huang et al., 2007; Lai et al., 2010; Xu et al., 2015). In this study, indoor $PM_{2.5}$ in residential homes in four Chinese cities (i.e., Hong Kong (HK), Guangzhou (GZ), Shanghai (SH), and Xi'an (XA)) with different pollution mixtures, meteorological characteristics, and residential living habits were for the first time investigated during the same sampling period from November 2016 to April 2017, for assessing the characteristics of potential sources, and examining the spatial variation of source contributions.

2 Methods

2.1 Sampling sites

Households in four Chinese cities were investigated in this study. Hong Kong is a well-developed coastal city located in south China with high population and urban density. Major air pollutant emission sources in Hong Kong are ocean cargo carriers, road traffic, and domestic emissions. Guangzhou is the capital city of Guangdong province in south China relying on its commercial and moderate industrial activities, with ship emission, traffic emission and industrial emission as the major sources of air pollution. Shanghai is the largest city located at the estuary of Yangtze River Delta, for which ship emission and traffic emission are dominated. Xi'an is an inland city located at the centre of Guanzhong Plain in west-northern part of China, relying on its industrial and tourism activities. Sources of the air pollutants during winter in Xi'an include industrial emission, traffic emission, and from heating services. Locations of the four cities are shown in Fig. 1. According to the long-term record from National Oceanic and Atmospheric Administration (NOAA) of USA, the average ambient air temperature during winter for Hong Kong, Guangzhou, Shanghai, and Xi'an are 15 – 17 ºC, 15 – 17 ºC, 5 – 7 ºC, and 1 – 3 ºC, respectively.

A total of 68 residential homes, with two sets of indoor $PM_{2.5}$ samples collected for most of the residences, were investigated for this study. Sampling periods were 9th November 2016 – 8th March 2017 (39 days) for Hong Kong, 29th November 2016 – 15th March 2017 (14 days) for Guangzhou, 21st November 2016 – 14th April 2017 (37 days) for Shanghai, and 18th November 2016 – 20th March 2017 (41 days) for Xi'an, respectively. In terms of urban planning, Shanghai and Xi'an share similar strategy, that residential areas are concentratedly located in suburban area with less commercial infrastructures, while urban areas are mainly for commercial activities with less and separated residential areas. Hence, most of the residences investigated in Shanghai (13 cases, 65.7%) and Xi'an (18 cases, 85.7%) investigated in this study were classified as suburban residences. While Guangzhou was with less centralized planning, that the residential areas are located more separately with commercial infrastructures nearby in the city centre areas (Liwan, Yuexiu, Haizhu, and Tianhe Districts). Four out of seven residences investigated in Guangzhou were identified as urban residences (57.1%) as they located in city centre areas, while the other three located in districts far from centre areas and with less commercial infrastructures nearby were identified as suburban residences (42.9%). A large number of population reside in Hong Kong Island, Kowloon district, and along the train line in New Territories district, where residential areas are closely combined with commercial infrastructures. Hence, most of the investigated homes were identified as urban residences (85.0%) in Hong Kong. Information including location categories, elevation, floor area, number of residents, and indoor activities is summarized in Table S1. As shown in Fig. S1, most of the residences investigated in this study were separately located in districts in city centre areas and centralized residential areas in suburban, with some located in districts far from city centre (suburban or even rural areas). Moreover, the investigated residences include different normal family types (with or without indoor cooking, tobacco smoking activities, different size of home occupants, etc.). We considered the residences investigated in this study were capable to represent the general condition of the city.

2.2 Sample Collection

Simultaneous $PM_{2.5}$ sampling with 47 mm Teflon-membrane filters and Quartz-fibre filters (PALL, USA) were conducted using two MiniVol Portable Air Samplers (Airmetrics, USA) equipped with $PM_{2.5}$ impactors with a flow rate of 5 L min^{-1} at a sampling height of 1.5 m above the ground level. The sampling period between November 2016 and early April 2017 intends to represent winter and early spring seasons. Concurrent real-time monitoring of 1-minute average carbon dioxide (CO_2), temperature, and relative humidity by Q-trak (TSI, USA) were used to estimate the air exchanges, and 1-minute average $PM_{2.5}$ mass by Dust-trak (TSI, USA) was used to record temporal variations of indoor $PM_{2.5}$ level. Two consecutive 24-hour samples were acquired at most of the residential homes. Sampling log sheets were distributed for recording sampling information and the indoor activities.

Most of the outdoor $PM_{2.5}$ samples were collected at the balconies of residential homes. Due to the limited accessibility for outdoor sampling at some households, $PM_{2.5}$ measurements from nearby monitoring stations were applied to examine indoor and outdoor relationships. This includes $PM_{2.5}$ data from Hong Kong Environmental Protection Department (Hong Kong Environmental Protection Department), and data centre of Ministry of Ecology and Environment of the People’s Republic of China (Ministry of Ecology and Environment of the People’s Republic of China).

2.3 Sample processing

Teflon-membrane filters were equilibrated in a temperature (25.0 ± 1.0 °C) and relative humidity (40.0 ± 1.0 %) controlled environment for a minimum of 24 hours before the gravimetric analysis. $PM_{2.5}$ mass concentrations were determined by a microbalance with sensitivity of ± 1 µg (Sartorius, MC5, Germany). Consecutive weighing with interval of at least 24 hours were conducted until the mass difference between the two weighing is less than ± 15 µg. Average of the two weighing was used for reporting the $PM_{2.5}$ mass (Cao et al., 2005; Gao et al., 2015).

Quartz-fibre filters were pre-heated at 900 °C for 3 hours to remove the gaseous OC contaminants prior to sample collection. After sample collection, a DRI Model 2001 Thermal/Optical Carbon Analyzer (AtmAA Inc. Calabasas, CA, USA) was employed for carbon analysis of quartz-fibre filter samples, following the IMPROVEA thermal/optical reflectance

(TOR) protocol (Chow et al., 2007). A punch (0.526 cm^2) of the quartz-fibre filter was heated stepwise to 140 °C (OC1), 280 °C (OC2), 480 °C (OC3), and 580 °C (OC4) in a non-oxidizing helium atmosphere, then continuously heated in an oxidizing 2% oxygen with helium balance atmosphere at 580 °C (EC1), 740 °C (EC2), and 840 °C (EC3). Pyrolysis of OC is continuously monitored by a 632.8 nm wavelength helium-neon (He-Ne) laser. The evolved carbon is oxidized to CO_2 and then reduced to methane (CH_4) for quantification by a flame ionization detector (FID). The method detection limits (MDL) are 0.45 μg cm^{-2} for OC, 0.06 μg cm^{-2} for EC, and 0.45 μg cm^{-2} for total carbon (TC). All samples in this study yield concentrations higher than the MDL. Routine calibrations of twice a day during sample analysis were conducted, by injecting known quantity of CH_4 into the analyser for analysis.

$PM_{2.5}$ samples were stored in petri slides (Millipore, USA) under low temperature (i.e., < 4 °C) to minimize evaporation of organic compounds. For consideration of quality assurance and control (QA/QC), a total of 15 pairs of filters were collected and analysed as filed blank samples (Cao et al., 2003). Clean filters as well as the samples collected may be affected during the storage, transportation, and operation processes due to exposed to ambient air, that gas-phase organic compounds may be adsorbed onto the filters or samples. Mass change of field blank filters (2.1 μg $filter^{-1}$ in this study) was subtracted for the correction of $PM_{2.5}$ mass, and OC mass detected (0.45 μg cm^{-2} in this study, EC determined from field blanks were below the detection limit) was subtracted for correction of OC mass. A part of the OC/EC analysis results of the collected samples were invalid. Real-time monitoring instruments and mini-volume samplers were maintained and calibrated prior to sampling campaign by the Laboratory of the Hong Kong Polytechnic University and the State Key Laboratory of Aerosol Chemistry and Physics, Chinese Academy of Sciences in Xi'an.

2.4 Data analysis

Using CO_2 concentrations to estimate air exchange rate for individual households assumes that: 1) no indoor CO_2 source after the residents left home, and 2) the indoor CO_2 concentration decreases logarithmically, which can be expressed by:

$$C_t = C_a \times (1 - e^{-It}) + C_0 \times e^{-It} \quad (1)$$

$$\ln(C_t - C_a) = -It + \ln(C_0 - C_a) \quad (2)$$

Where C_0 is the initial indoor CO_2 concentration; C_t is the CO_2 concentration at time t; C_a is the ambient CO_2 concentration; and I is the air exchange rate in h^{-1}, which can be obtained as the slope of the fitting curve of Eq. (2) derived from Eq. (1). Similar tracer gas decay method has been reported for evaluating the ventilation conditions in residential premises (Chao et al., 1998).

3 Results and discussion

3.1 Mass concentrations of $PM_{2.5}$ and organic/elemental carbon

Statistical summary of average indoor and outdoor $PM_{2.5}$ concentrations in each city is shown in Table 1. The lowest average indoor (34.0 ± 14.6 μg m^{-3}) and corresponding outdoor (32.9 ± 12.6 μg m^{-3}) $PM_{2.5}$ concentrations were found for Hong Kong households, while residents in Xi'an were exposed to the highest indoor (78.7 ± 49.3 μg m^{-3}) and outdoor (109.3 ± 64.4 μg m^{-3}) concentrations. During the sampling period of 41 days in Xi'an, ambient $PM_{2.5}$ concentrations exceeded the Chinese *Heavily Polluted* standard of 150 μg m^{-3} on 10 days (24.4% of the period), and among which 2 days (4.9%) exceeded the *Severe Polluted* standard of 250 μg m^{-3} (Inspection and Quarantine of the People's Republic of China, 2012). While average indoor $PM_{2.5}$

concentration in Shanghai (50.3 ± 17.9 μg m^{-3}) and Guangzhou (47.2 ± 5.4 μg m^{-3}) are comparative to Interim Target 2 (IT-2) of WHO Air Quality Guidance of 50 μg m^{-3}, average concentration in Hong Kong (34.0 ± 14.6 μg m^{-3}) are lower than the IT-3 of 37.5 μg m^{-3}. Average indoor $PM_{2.5}$ in Xi'an (78.7 ± 49.3 μg m^{-3}) exceeded WHO's IT-1 level of 70 μg m^{-3}, suggesting the severity of indoor air pollution. As illustrated in Table 2, indoor $PM_{2.5}$ concentrations in this study are obviously lower than those of previous studies (Cao et al., 2012; Chao and Wong, 2002; Ho et al., 2004; Lai et al., 2010; Zhou et al., 2018; Zhu et al., 2012).

For the associated organic/elemental carbon levels, Table 3 shows highest indoor OC (23. 5 ± 9.9 μg m^{-3}) and EC (6. 7 ± 4.0 μg m^{-3}) concentrations in Xi'an with comparable OC (12.9 – 15.8 μg m^{-3}) and EC (3.1 – 3.7 μg m^{-3}) in other cities. Table 4 shows carbonaceous aerosol, i.e., organic matter (OM) + EC, accounts for 48.9 – 62.8 % and 40.7 – 50.4 % of indoor and outdoor $PM_{2.5}$ mass, respectively. High carbon fractions were identified in both indoor (38.5 ± 7.8 % for OC and 8.9 ± 2.3 % for EC) and outdoor (30.2 ± 9.1 % for OC and 8.2 ± 4.0 % for EC) samples in Hong Kong. Hong Kong is a dense city with less distance between buildings compared with other cities, thus the influence on indoor PM from traffic emission, which is enriched in carbon species could be significant (Cao et al., 2003; Cao et al., 2004; Ho et al., 2003). Larger difference between indoor and outdoor OC (2.6 – 8.4 %) than EC (-2.2 – 1.5 %) suggesting the presence of indoor OC sources. The OC to $PM_{2.5}$ ratio (30 – 39 %) and OM to $PM_{2.5}$ ratio (41 – 54 %) from this study is about 8 % and 11 % higher than the previous studies (22 – 29 % for OC, and 31 – 41 % for OM) (Cao et al., 2012; Cao et al., 2005; Ho et al., 2004; Lai et al., 2010; Zhu et al., 2012) for indoor $PM_{2.5}$.

The OC/EC ratio has been used to characterizing the emission and transformation of carbonaceous aerosol and to indicate the presences of secondary organic carbon (SOC) (Gray et al., 1986). Cao et al. (2007) reported the critical OC/EC ratio values of SOC presence during winter were 2.81 and 2.13 for northern and southern Chinese cities, respectively. Table 4 shows high OC/EC ratios, ranging from 3.9 to 4.5 for indoor and from 2.8 to 4.2 for outdoor, which were all higher than the reference levels (Cao et al., 2007), implying the presence of SOC for both indoors and outdoors for all cities. Higher OC/EC ratios for indoor than outdoor and good indoor OC-EC correlations (r = 0.8 – 0.9) also confirmed the presence of indoor combustion sources of carbonaceous matters.

3.2 Effects of air exchange

The relationship between the indoor and outdoor air quality is complex, the indoor to outdoor (I/O) ratio and indoor-outdoor correlation of $PM_{2.5}$ concentration, as well as air exchange rate (AER) are commonly applied to evaluate the degree of penetration from outdoor to indoor. Statistical summary of $PM_{2.5}$ I/O ratio and the AER of residential homes are shown in Table 1, with indoor/outdoor $PM_{2.5}$ correlations in Table 1.

The lowest average $PM_{2.5}$ I/O ratio (0.72 ± 0.23) and AER (0.8 ± 0.8 h^{-1}), as well as the highest indoor $PM_{2.5}$ level (78.7 ± 49.3 μg m^{-3}) were found for Xi'an households with highest indoor-outdoor correlation (r = 0.89). This is attributed to the restricted air exchange for maximizing the heating efficiency during centralized heating service period in Xi'an, which is not applied in other cities with warmer climate. Highly correlated indoor and outdoor $PM_{2.5}$ levels in Xi'an households suggest the limited contribution from indoor sources, also the high outdoor $PM_{2.5}$ levels may have influence on indoor levels to some extent. During cold days, residences in Shanghai tend to use residential HVAC (heating, ventilation, and air conditioning) system. This resulted in a moderate level of AER (1.3 ± 1.1 h^{-1}) in Shanghai residences. With considerably low outdoor $PM_{2.5}$ levels (38.2 ± 18.8 μg m^{-3}), the effect of outdoor penetration may be limited for Shanghai households. The highest I/O ratio (1.32 ± 0.43) for Shanghai households (Table 1) implies significant contribution from indoor sources. Residences in Hong Kong experienced the highest AER of 4.3 ± 3.4 h^{-1} and moderate I/O ratio of 1.03 ± 0.40, suggesting the influence of outdoor penetration, but with poor indoor-outdoor correlation (r = 0.52, p < 0.05). These results are consistent with those reported by Chao and Wong (2002) with I/O ratio of 0.96, AER of 4.6 h^{-1}, and indoor-outdoor correlation (r < 0.26, p = 0.14) for 34 Hong Kong households. As most of the households in Hong Kong are apparently smaller in interior volume, the effects of outdoor penetration or indoor activities could be sensitively reflected on indoor air quality.

Relationships between indoor and outdoor $PM_{2.5}$, as well as the portions of carbonaceous species in indoor $PM_{2.5}$ mass during different degrees of pollution were thus investigated. According to the Technical Regulation on Ambient Air Quality Index of China (Ministry of Environmental Protection of the People's Republic of China, 2012), the obtained samples with 24-hr sampling period were classified into four groups on the basis of corresponding outdoor $PM_{2.5}$ concentrations: < 35.0 µg m^{-3} (Clear), 35.1 ~ 75.0 µg m^{-3} (Light Pollution), 75.1 ~ 115.0 µg m^{-3} (Medium Pollution), and > 115 µg m^{-3} (Heavy Pollution). Samples collected in Hong Kong, Guangzhou, and Shanghai were mostly during Clear days (N=41) and Light Pollution days (N = 54), while for most of the samples collected during Medium Pollution days (N = 23) and Heavy Pollution days (N = 11) were from Xi'an. A descending trend of $PM_{2.5}$ I/O ratio (from 1.38 for Clear days to 0.73 for Heavy Pollution days) could be observed as the worsening ambient air quality in Fig. 2a. Distribution of I/O ratio of individual homes on various outdoor $PM_{2.5}$ concentration is shown in Fig. 2b, that higher I/O ratio cases were mostly identified during Clear days and Light Pollution periods, while most cases during Heavy Pollution days were with I/O ratios lower than unity. This distribution indicates the strong influence from outdoor penetration during air pollution episodes, while contribution from indoor sources were likely to be prominent during periods with good air quality. Fig. 3 shows the portions in $PM_{2.5}$ mass of the carbonaceous fractions during different degrees of air pollution. A descending trend could also be observed of the TC portions in $PM_{2.5}$ mass. No significant difference was observed for EC portions (8.1 % ~ 8.7 %), while OC portions showed a 7.4 % difference from Clear days (35.4 %) to Heavy Pollution days (28.0 %). As previously discussed, OC portions in outdoor $PM_{2.5}$ were generally lower than indoor $PM_{2.5}$ since the existence of typical indoor OC sources such as cooking and tobacco smoking. Regardless the limited air exchange identified for Xi'an households, most of the samples were collected during poor air quality period, that lower $PM_{2.5}$ I/O ratios and indoor OC portions for these Xi'an households could be attributed to more significant influence from outdoor penetration.

3.3 Effects of indoor activities

According to different indoor activities (e.g. cooking, smoking, and incense burning), households in four cities were classified into three types:

- Type A: no specific combustion activity (i.e., cooking, tobacco smoking, or incense burning);
- Type B: only with cooking activities; and
- Type C: with cooking and other combustion activities (e.g., tobacco smoking and/or incense burning).

Majority of the residential homes for this study were classified as Type B (60.0%, 57.1%, 50.0%, and 61.9% of investigated homes in Hong Kong, Guangzhou, Shanghai, and Xi'an, respectively) and Type C (20.0%, 42.9%, 35.0%, and 38.1% in Hong Kong, Guangzhou, Shanghai, and Xi'an), with limited Type A households (15.0% and 10.0% in Hong Kong and Shanghai). Two households with indoor tobacco smoking but with no cooking were excluded.

Table 5 shows a general trend of Type C > Type B > Type A for indoor $PM_{2.5}$ for four cities. Considering only the residences of Hong Kong and Shanghai, average $PM_{2.5}$ of Type B (37.6 ± 14.7 μg m^{-3}) and Type C (51.0 ± 20.0 μg m^{-3}) households were 54.1% and 109.0% higher than those of Type A households (24.4 ± 13.0 μg m^{-3}), respectively. With relatively less variations in outdoor $PM_{2.5}$ concentrations for these two cities, a trend of $PM_{2.5}$ I/O ratios of Type C (1.42) > Type B (1.04) > Type A (0.92) were found, confirming the impact from indoor combustion sources. On average for four cities, the indoor $PM_{2.5}$ for Type C households was 23.1% higher than those of Type B, showing the additional impacts from tobacco smoking and/or incense burning. This is consistent with the findings that indoor tobacco smoking could induce a 18 % increase of indoor $PM_{2.5}$ reported by Chao and Wong (2002). With similar outdoor $PM_{2.5}$ levels, the I/O ratios increased from 0.92 to 1.30 for Hong Kong and from 1.19 to 1.49 for Shanghai from Type B to Type C households, and with 37.1% and 26.5% increments in $PM_{2.5}$ concentrations, respectively. Relatively small differences in $PM_{2.5}$ were found between Types B and C households in Guangzhou and Xi'an. With elevated $PM_{2.5}$ concentrations in Xi'an, the atypical relationship between the I/O ratios were found for Type B (0.73) and Type C (0.71) households with high indoor-outdoor correlation (r = 0.94 and 0.88, respectively), confirming the limited influence induced by indoor combustion activities on the indoor $PM_{2.5}$ levels for Xi'an households.

Table 6 shows that in general, increments of 7.2 μg m^{-3} (79.9 %) for OC and 1.7 μg m^{-3} (69.0 %) for EC from Type A to Type B household, and increments of 3.0 μg m^{-3} (18.7 %) for OC and 1.2 μg m^{-3} (30.3 %) for EC from Type B to Type C household were identified. The exception is found for Shanghai where OC and EC were found higher for Type B (14.9 ± 6.0 μg m^{-3}) than Type C (13.6 ± 2.8 μg m^{-3}) households. Tobacco smoking and/or incense burning may represent smoldering combustion that yields higher OC/EC ratios. Previous studies (Cao et al., 2005; Lai et al., 2010) reported average OC/EC ratios of around 10 of the households with intensive indoor tobacco smoking and incense burning activities, with the highest OC/EC ratio of 21.2 (around 5-fold of results in this study). Correlations between indoor OC and EC for different home types were also studied, as shown in Table 7. Better correlation for Type B (r = 0.95) compared with Type C households (r = 0.81) indicates the presence of multiple OC sources for Type C households other than indoor combustions.

Selected examples of time-series plot of 10-minute average indoor $PM_{2.5}$ concentrations are illustrated in Fig. 4. Without indoor activity, Fig. 4a shows little fluctuations in $PM_{2.5}$ concentrations for Type A compared with Type B and C households. This diurnal pattern showed an elevated indoor $PM_{2.5}$ concentration during the daytime (up to about 60 µg m^{-3}) and lowering during night-time to about 15 µg m^{-3}, which may in accordance with the pattern of outdoor $PM_{2.5}$ levels. Considerably higher background $PM_{2.5}$ concentration of ~ 90 µg m^{-3} were identified for this Type B household (Fig. 4b), with apparent spikes (> 100 µg m^{-3}, about 190 – 470% higher than background) during meal preparation time (e.g., 06:00 and 18:00 LST). Fig. 4c shows more apparent spikes (6 to 26-fold) with the maximum 10-minute average $PM_{2.5}$ concentrations exceeding 2000 µg m^{-3} (around the noon on 29th November 2016). Considering there was no outdoor source other than traffic emission as HK-05 is located next to a main road with heavy traffic, the rapid increases observed for indoor $PM_{2.5}$ concentration were not likely to be attributed to outdoor penetration. Routine incense burning as religious practice, and indoor cooking activities according to sampling log, were the main contributors to the spikes observed for this household.

A simple equation was developed for further understand the PM source contributions, generally for households in Hong Kong and Shanghai.

$$C_{indoor} = C_{outdoor-penetrate} + C_{cooking} + C_{smoking/incense-burning} + C_{indoor-other} \quad (3)$$

Where $C_{outdoor\text{-}penetrate}$ is the mean outdoor $PM_{2.5}$ concentration of Type A households; $C_{cooking}$ is the corrected $PM_{2.5}$ concentrations contributed by cooking activities based on the differences between the average Type B and Type A households; $C_{smoking/incense\ burning}$ is corrected concentration contributed by tobacco smoking and/or incense burning based on the differences between Type C and Type B household; and $C_{indoor\ other}$ is the corrected differences between average indoor and outdoor concentrations of Type A households, which may be contributed by other indoor sources such as resuspension of deposited dust due to the movement of occupants. Nine homes in Hong Kong (2 Type A, 4 Type B, and 3 Type C homes) and eight homes in Shanghai (1 Type A, 4 Type B, and 3 Type C homes) with similar outdoor $PM_{2.5}$ levels were selected for this equation analysis for avoid excessive influence of outdoor penetration. Correction was performed based on the difference of average outdoor $PM_{2.5}$ concentration.

Fig. 5 shows that with the highest AER ($4.3 \pm 3.4\ h^{-1}$, Table 1) in Hong Kong, over half of the $PM_{2.5}$ (57.4%) is attributed to outdoor penetration, much higher than the 43.2% for Shanghai (AER of $1.3 \pm 1.1\ h^{-1}$). This difference is also reflected by the difference between outdoor and indoor concentrations (i.e., $C_{indoor\ other}$ in Eq. 3 with 4-fold higher contribution for Shanghai). While cooking activities contributes similar (15.6 %) portions for both cities. Smoking and/or incense burning contributions are higher for Hong Kong (20.4 %) than Shanghai (12.7 %).

4 Conclusions

The indoor $PM_{2.5}$ and associated OC and EC were examined in 68 residential homes in four Chinese cities during the period from November 2016 to early April 2017. A general trend of Xi'an > Shanghai > Guangzhou > Hong Kong for indoor $PM_{2.5}$ levels was identified. Descending trend of I/O ratio as increase of outdoor $PM_{2.5}$ level suggested the indoor air quality could be influenced by outdoor penetration during air pollution episodes. Highest indoor OC and EC levels were identified in Xi'an's households, while those in other three cities were with similar levels. OM accounted larger portion to indoor $PM_{2.5}$ when compared with previous studies, with high OC/EC ratio suggesting the presence of SOC in indoors for the four cities. By dividing the cases into three categories according to indoor activities, apparent increment of indoor $PM_{2.5}$ and associated OC concentration were observed with more indoor combustion sources. Large difference of OC in indoor and outdoor indicated the presence of indoor OC sources. From the proposed equation for estimating the contribution from different sources to indoor $PM_{2.5}$ levels, contribution from outdoor penetration was found predominant for Hong Kong households which were with high level of air exchange, while the indoor sources other than cooking, tobacco smoking or incense burning accounted considerable portion of contribution for Shanghai households.

With the worst ambient air quality during sampling period among the four cities, indoor air quality of Xi'an households was found significantly affected by outdoor penetration. Contribution from indoor sources was identified predominant for Shanghai households, indicated by the highest $PM_{2.5}$ I/O ratio. Though households in Hong Kong were with higher level of air exchange than other cities, the influence of indoor sources was identified significant, leading to a $PM_{2.5}$ I/O ratio close to unity and poor correlation between indoor and outdoor $PM_{2.5}$ concentration. Overall, for lowering health risks associated with particle inhalation, indoor air quality may need further improved for Chinese households, especially for the period of worse ambient air quality.

Acknowledgements

This study was supported by the Research Grants Council of Hong Kong Government (Project No. PolyU 152090/15E), the Research Grants Council of Hong Kong Government (Project No. T24/504/17), and the National Key Research and Development Program of China (2016YFA0203000). The authors would also like to thank our previous research group member, Mr. LAI Nguyen Huy, the homeowners, and research group members from South China University of Technology, East China University of Science and Technology, and Institute of Earth Environment, Chinese Academy of Sciences for cooperation on indoor sampling.

References

Abdullahi, K.L., Delgado-Saborit, J.M., Harrison, R.M. (2013). Emissions and indoor concentrations of particulate matter and its specific chemical components from cooking: A review. Atmos. Environ. 71, 260-294. https://doi.org/10.1016/j.atmosenv.2013.01.061.

Abt, E., Suh, H.H., Catalano, P., Koutrakis, P. (2000). Relative Contribution of Outdoor and Indoor Particle Sources to Indoor Concentrations. Environ. Sci. Technol. 34, 3579-3587. https://doi.org/10.1021/es990348y.

Achilleos, S., Kioumourtzoglou, M.-A., Wu, C.-D., Schwartz, J.D., Koutrakis, P., Papatheodorou, S.I. (2017). Acute effects of fine particulate matter constituents on mortality: A systematic review and meta-regression analysis. Environ. Int. 109, 89-100. https://doi.org/10.1016/j.envint.2017.09.010.

Barraza, F., Jorquera, H., Valdivia, G., Montoya, L.D. (2014). Indoor $PM_{2.5}$ in Santiago, Chile, spring 2012: Source apportionment and outdoor contributions. Atmos. Environ. 94, 692-700. https://doi.org/10.1016/j.atmosenv.2014.06.014.

Cao, J.-J., Zhu, C.-S., Tie, X.-X., Geng, F.-H., Xu, H.-M., Ho, S.S.H., Wang, G.-H., Han, Y.-M., Ho, K.-F. (2013). Characteristics and sources of carbonaceous aerosols from Shanghai, China. Atmos. Chem. Phys. 13, 803-817. https://doi.org/10.5194/acp-13-803-2013.

Cao, J.J., Huang, H., Lee, S.C., Chow, J.C., Zou, C.W., Ho, K.F., Watson, J.G. (2012). Indoor/Outdoor Relationships for Organic and Elemental Carbon in $PM_{2.5}$ at Residential Homes in Guangzhou, China. Aerosol Air Qual. Res. 12, 902-910. https://doi.org/10.4209/aaqr.2012.02.0026.

Cao, J.J., Lee, S.C., Chow, J.C., Cheng, Y., Ho, K.F., Fung, K., Liu, S.X., Watson, J.G. (2005). Indoor/outdoor relationships for $PM_{2.5}$ and associated carbonaceous pollutants at residential homes in Hong Kong - case study. Indoor Air 15, 197-204. https://doi.org/10.1111/j.1600-0668.2005.00336.x.

Cao, J.J., Lee, S.C., Chow, J.C., Watson, J.G., Ho, K.F., Zhang, R.J., Jin, Z.D., Shen, Z.X., Chen, G.C., Kang, Y.M., Zou, S.C., Zhang, L.Z., Qi, S.H., Dai, M.H., Cheng, Y., Hu, K. (2007). Spatial and seasonal distributions of carbonaceous aerosols over China. J. Geophys. Res. 112, D22S11. https://doi.org/10.1029/2006jd008205.

Cao, J.J., Lee, S.C., Ho, K.F., Zhang, X.Y., Zou, S.C., Fung, K., Chow, J.C., Watson, J.G. (2003). Characteristics of carbonaceous aerosol in Pearl River Delta Region, China during 2001 winter period. Atmos. Environ. 37, 1451-1460. https://doi.org/10.1016/S1352-2310(02)01002-6.

Cao, J.J., Lee, S.C., Ho, K.F., Zou, S.C., Fung, K., Li, Y., Watson, J.G., Chow, J.C. (2004). Spatial and seasonal variations of atmospheric organic carbon and elemental carbon in Pearl River Delta Region, China. Atmos. Environ. 38, 4447-4456. https://doi.org/10.1016/j.atmosenv.2004.05.016.

Chao, C.Y., Wong, K.K. (2002). Residential indoor PM_{10} and $PM_{2.5}$ in Hong Kong and the elemental composition. Atmos. Environ. 36, 265-277. https://doi.org/10.1016/S1352-2310(01)00411-3.

Chao, C.Y.H., Tung, T.C.W., Burnett, J. (1998). Influence of Different Indoor Activities on the Indoor Particulate Levels in Residential Buildings. Indoor Built Environ. 7, 110-121. https://doi.org/10.1159/000024569.

Chen, R., Zhao, Z., Kan, H. (2013). Heavy Smog and Hospital Visits in Beijing, China. Am. J. Resp. Crit. Care 188, 1170-1171. https://doi.org/10.1164/rccm.201304-0678LE.

Cheng, X., Zhang, H., Pan, W., Liu, S., Zhang, M., Long, Z., Zhang, T., Chen, Q. (2017). Field study of infiltration rate and its influence on indoor air quality in an apartment. Procedia Engineer. 205, 3954-3961. https://doi.org/10.1016/j.proeng.2017.09.853.

Chow, J.C., Watson, J.G., Chen, L.-W.A., Chang, M.C.O., Robinson, N.F., Trimble, D., Kohl, S. (2007). The IMPROVE_A Temperature Protocol for Thermal/Optical Carbon Analysis: Maintaining Consistency with a Long-Term Database. J. Air Waste Manage. Assoc. 57, 1014-1023. https://doi.org/10.3155/1047-3289.57.9.1014.

Custódio, D., Pinho, I., Cerqueira, M., Nunes, T., Pio, C. (2014). Indoor and outdoor suspended particulate matter and associated carbonaceous species at residential homes in northwestern Portugal. Sci. Total Environ. 473-474, 72-76. https://doi.org/10.1016/j.scitotenv.2013.12.009.

Eatough, D.J., Benner, C.L., Tang, H., Landon, V., Richards, G., Caka, F.M., Crawford, J., Lewis, E.A., Hansen, L.D., Eatough, N.L. (1989). The chemical composition of environmental tobacco smoke III. Identification of conservative tracers of environmental tobacco smoke. Environ. Int. 15, 19-28. https://doi.org/10.1016/0160-4120(89)90005-6.

Frazer, L. (2002). Seeing through soot. Environ. Health Perspect. 110, A470-A473. https://doi.org/10.1289/ehp.110-a470.

Funasaka, K., Miyazaki, T., Tsuruho, K., Tamura, K., Mizuno, T., Kuroda, K. (2000). Relationship between indoor and outdoor carbonaceous particulates in roadside households. Environ. Pollut. 110, 127-134. https://doi.org/10.1016/S0269-7491(99)00281-X.

Gao, Y., Lai, S., Lee, S.-C., Yau, P.S., Huang, Y., Cheng, Y., Wang, T., Xu, Z., Yuan, C., Zhang, Y. (2015). Optical properties of size-resolved particles at a Hong Kong urban site during winter. Atmos. Res. 155, 1-12. https://doi.org/10.1016/j.atmosres.2014.10.020.

Gray, H.A., Cass, G.R., Huntzicker, J.J., Heyerdahl, E.K., Rau, J.A. (1986). Characteristics of atmospheric organic and elemental carbon particle concentrations in Los Angeles. Environ. Sci. Technol. 20, 580-589. https://doi.org/10.1021/es00148a006.

He, L.-Y., Hu, M., Huang, X.-F., Yu, B.-D., Zhang, Y.-H., Liu, D.-Q. (2004). Measurement of emissions of fine particulate organic matter from Chinese cooking. Atmos. Environ. 38, 6557-6564. https://doi.org/10.1016/j.atmosenv.2004.08.034.

Heo, J., Schauer, J.J., Yi, O., Paek, D., Kim, H., Yi, S.-M. (2014). Fine particle air pollution and mortality: importance of specific sources and chemical species. Epidemiology 25, 379-388. https://doi.org/10.1097/EDE.0000000000000044.

Ho, K.F., Cao, J.J., Harrison, R.M., Lee, S.C., Bau, K.K. (2004). Indoor/outdoor relationships of organic carbon (OC) and elemental carbon (EC) in $PM_{2.5}$ in roadside environment of Hong Kong. Atmos. Environ. 38, 6327-6335. https://doi.org/10.1016/j.atmosenv.2004.08.007.

Ho, K.F., Lee, S.C., Chan, C.K., Yu, J.C., Chow, J.C., Yao, X.H. (2003). Characterization of chemical species in PM2.5 and PM10 aerosols in Hong Kong. Atmos. Environ. 37, 31-39. https://doi.org/10.1016/S1352-2310(02)00804-X.

Ho, K.F., Lee, S.C., Yu, J.C., Zou, S.C., Fung, K. (2002). Carbonaceous characteristics of atmospheric particulate matter in Hong Kong. Sci. Total Environ. 300, 59-67. https://doi.org/10.1016/S0048-9697(02)00281-4.

Hong Kong Environmental Protection Department. Inquire and Download Air Quality Monitoring Data. 2019. https://cd.epic.epd.gov.hk/EPICDI/air/station/.

Huang, H., Lee, S.-C., Cao, J.-J., Zou, C.-W., Chen, X.-G., Fan, S.-J. (2007). Characteristics of indoor/outdoor $PM_{2.5}$ and elemental components in generic urban, roadside and industrial plant areas of Guangzhou City, China. J. Environ. Sci. 19, 35-43. https://doi.org/10.1016/s1001-0742(07)60006-0.

Huang, R.-J., Zhang, Y., Bozzetti, C., Ho, K.-F., Cao, J.-J., Han, Y., Daellenbach, K.R., Slowik, J.G., Platt, S.M., Canonaco, F., Zotter, P., Wolf, R., Pieber, S.M., Bruns, E.A., Crippa, M., Ciarelli, G., Piazzalunga, A., Schwikowski, M., Abbaszade, G., Schnelle-Kreis, J., Zimmermann, R., An, Z., Szidat, S., Baltensperger, U., El Haddad, I., Prévôt, A.S.H. (2014). High secondary aerosol contribution to particulate pollution during haze events in China. Nature 514, 218-222. https://doi.org/10.1038/nature13774.

Inspection and Quarantine of the People's Republic of China. Ambient Air Quality Standards (GB 3095-2012), 2012. http://kjs.mee.gov.cn/hjbhbz/bzwb/dqhjbh/dqhjzlbz/201203/t20120302_224165.shtml.

Klepeis, N.E., Nelson, W.C., Ott, W.R., Robinson, J.P., Tsang, A.M., Switzer, P., Behar, J.V., Hern, S.C., Engelmann, W.H. (2001). The National Human Activity Pattern Survey (NHAPS): a resource for assessing exposure to environmental pollutants. J. Expo. Sci. Env. Epidemiol. 11, 231-252. https://doi.org/10.1038/sj.jea.7500165.

Lai, S.C., Ho, K.F., Zhang, Y.Y., Lee, S.C., Huang, Y., Zou, S.C. (2010). Characteristics of Residential Indoor Carbonaceous Aerosols: A Case Study in Guangzhou, Pearl River Delta Region. Aerosol Air Qual. Res. 10, 472-478. https://doi.org/10.4209/aaqr.2010.05.0037.

Lee, S.-C., Wang, B. (2004). Characteristics of emissions of air pollutants from burning of incense in a large environmental chamber. Atmos. Environ. 38, 941-951. https://doi.org/10.1016/j.atmosenv.2003.11.002.

Li, L., Ho, S.S.H., Chow, J.C., Watson, J.G., Lee, F.S.C., Cui, L., Gao, Y., Dai, W., Ho, K.F., Huang, Y., Cao, J. (2017). Characterization and health risk assessment of $PM_{2.5}$-bound organics inside and outside of Chinese smoking lounges. Chemosphere 186, 438-445. https://doi.org/10.1016/j.chemosphere.2017.08.006.

Liu, J., Mauzerall, D.L., Chen, Q., Zhang, Q., Song, Y., Peng, W., Klimont, Z., Qiu, X., Zhang, S., Hu, M., Lin, W., Smith, K.R., Zhu, T. (2016). Air pollutant emissions from Chinese households: A major and underappreciated ambient pollution source. Proc. Natl. Acad. Sci. U.S.A. 113, 7756-7761. https://doi.org/10.1073/pnas.1604537113.

Ministry of Ecology and Environment of the People's Republic of China. National Air Quality Data Publication Platform. http://www.mee.gov.cn/hjzl/.

Ministry of Environmental Protection of the People's Republic of China. Technical Regulation on Ambient Air Quality Index (on trial). HJ 633-2012, 2012. http://www.mee.gov.cn/ywgz/fgbz/bz/bzwb/jcffbz/201203/W020120410332725219541.pdf.

Morawska, L., He, C., Hitchins, J., Gilbert, D., Parappukkaran, S. (2001). The relationship between indoor and outdoor airborne particles in the residential environment. Atmos. Environ. 35, 3463-3473. https://doi.org/10.1016/S1352-2310(01)00097-8.

Moriske, H.-J., Drews, M., Ebert, G., Menk, G., Scheller, C., Schöndube, M., Konieczny, L. (1996). Indoor air pollution by different heating systems: coal burning, open fireplace and central heating. Toxicol. Lett. 88, 349-354. https://doi.org/10.1016/0378-4274(96)03760-5.

Nunes, T.V., Pio, C.A. (1993). Carbonaceous aerosols in industrial and coastal atmospheres. Atmos. Environ. A, Gen. Top. 27, 1339-1346. https://doi.org/10.1016/0960-1686(93)90259-2.

Patterson, E., Eatough, D.J. (2000). Indoor/Outdoor Relationships for Ambient $PM_{2.5}$ and Associated Pollutants: Epidemiological Implications in Lindon, Utah. J. Air Waste Manage. Assoc. 50, 103-110. https://doi.org/10.1080/10473289.2000.10463986.

Penttinen, P., Timonen, K.L., Tiittanen, P., Mirme, A., Ruuskanen, J., Pekkanen, J. (2001a). Number concentration and size of particles in urban air: effects on spirometric lung function in adult asthmatic subjects. Environ. Health Perspect. 109, 319-323. https://doi.org/10.1289/ehp.01109319.

Penttinen, P., Timonen, K.L., Tiittanen, P., Mirme, A., Ruuskanen, J., Pekkanen, J. (2001b). Ultrafine particles in urban air and respiratory health among adult asthmatics. Eur. Respir. J. 17, 428-435. https://doi.org/10.1183/09031936.01.17304280.

Pope III, C.A., Burnett, R.T., Thun, M.J., Calle, E.E., Krewski, D., Ito, K., Thurston, G.D. (2002). Lung Cancer, Cardiopulmonary Mortality, and Long-term Exposure to Fine Particulate Air Pollution. JAMA 287, 1132-1141. https://doi.org/10.1001/jama.287.9.1132.

Pope III, C.A., Dockery, D.W. (2006). Health Effects of Fine Particulate Air Pollution: Lines that Connect. J. Air Waste Manage. Assoc. 56, 709-742. https://doi.org/10.1080/10473289.2006.10464485.

Secrest, M.H., Schauer, J.J., Carter, E.M., Baumgartner, J. (2017). Particulate matter chemical component concentrations and sources in settings of household solid fuel use. Indoor Air 27, 1052-1066. https://doi.org/10.1111/ina.12389.

Shen, H., Tao, S., Chen, Y., Ciais, P., Güneralp, B., Ru, M., Zhong, Q., Yun, X., Zhu, X., Huang, T., Tao, W., Chen, Y., Li, B., Wang, X., Liu, W., Liu, J., Zhao, S. (2017). Urbanization-induced population migration has reduced ambient $PM_{2.5}$ concentrations in China. Sci. Adv. 3, e1700300. https://doi.org/10.1126/sciadv.1700300.

Tian, S.L., Pan, Y.P., Wang, Y.S. (2016). Size-resolved source apportionment of particulate matter in urban Beijing during haze and non-haze episodes. Atmos. Chem. Phys. 16, 1-19. https://doi.org/10.5194/acp-16-1-2016.

Wang, Z., Wang, T., Guo, J., Gao, R., Xue, L., Zhang, J., Zhou, Y., Zhou, X., Zhang, Q., Wang, W. (2012). Formation of secondary organic carbon and cloud impact on carbonaceous aerosols at Mount Tai, North China. Atmos. Environ. 46, 516-527. https://doi.org/10.1016/j.atmosenv.2011.08.019.

Xu, H., Guinot, B., Shen, Z., Ho, K.F., Niu, X., Xiao, S., Huang, R.-J., Cao, J. (2015). Characteristics of Organic and Elemental Carbon in $PM_{2.5}$ and $PM_{0.25}$ in Indoor and Outdoor Environments of a Middle School: Secondary Formation of Organic Carbon and Sources Identification. Atmosphere 6, 361-379. https://doi.org/10.3390/atmos6030361.

Zhang, X.Y., Wang, Y.Q., Niu, T., Zhang, X.C., Gong, S.L., Zhang, Y.M., Sun, J.Y. (2012). Atmospheric aerosol compositions in China: spatial/temporal variability, chemical signature, regional haze distribution and comparisons with global aerosols. Atmos. Chem. Phys. 12, 779-799. https://doi.org/10.5194/acp-12-779-2012.

Zhang, Y.-L., Cao, F. (2015). Fine particulate matter ($PM_{2.5}$) in China at a city level. Sci. Rep. 5, 14884. https://doi.org/10.1038/srep14884.

Zhou, S., Wang, T., Wang, Z., Li, W., Xu, Z., Wang, X., Yuan, C., Poon, C.N., Louie, P.K.K., Luk, C.W.Y., Wang, W. (2014). Photochemical evolution of organic aerosols observed in urban plumes from Hong Kong and the Pearl River Delta of China. Atmos. Environ. 88, 219-229. https://doi.org/10.1016/j.atmosenv.2014.01.032.

Zhou, S., Wang, Z., Gao, R., Xue, L., Yuan, C., Wang, T., Gao, X., Wang, X., Nie, W., Xu, Z., Zhang, Q., Wang, W. (2012). Formation of secondary organic carbon and long-range transport of carbonaceous aerosols at Mount Heng in South China. Atmos. Environ. 63, 203-212. https://doi.org/10.1016/j.atmosenv.2012.09.021.

Zhou, X., Cai, J., Zhao, Y., Chen, R., Wang, C., Zhao, A., Yang, C., Li, H., Liu, S., Cao, J., Kan, H., Xu, H. (2018). Estimation of residential fine particulate matter infiltration in Shanghai, China. Environ. Pollut. 233, 494-500. https://doi.org/10.1016/j.envpol.2017.10.054.

Zhu, C.-S., Cao, J.-J., Shen, Z.-X., Liu, S.-X., Zhang, T., Zhao, Z.-Z., Xu, H.-M., Zhang, E.-K. (2012). Indoor and Outdoor Chemical Components of $PM_{2.5}$ in the Rural Areas of Northwestern China. Aerosol Air Qual. Res. 12, 1157-1165. https://doi.org/10.4209/aaqr.2012.01.0003.

Declaration of competing interests

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.

Table Captions

Table 1. Statistical summary of indoor and outdoor $PM_{2.5}$ concentrations at four cities.

Table 2. Intercomparison of indoor $PM_{2.5}$ concentrations from different studies.

Table 3. Summary of indoor OC and EC concentrations and ranges in four cities.

Table 4. Percentage of OC, EC, and OM in $PM_{2.5}$ mass, OC-EC correlations in indoors and outdoors.

Table 5. Average indoor and outdoor $PM_{2.5}$ concentrations by three activity classifications.

Table 6. Average indoor $PM_{2.5}$ associated OC and EC concentrations by three activity classifications.

Table 1. Statistical summary of indoor and outdoor $PM_{2.5}$ concentrations at four cities.

	Indoor $PM_{2.5}$ Concentration (μg m^{-3})	Outdoor $PM_{2.5}$ Concentration (μg m^{-3})	I/O Ratio	Indoor-outdoor $PM_{2.5}$ Pearson Correlation Coefficient (r)	Air Exchange Rate (h^{-1})
Hong Kong (n=20) [a]	34.0 ± 14.6 (10.0 – 76.8) [b]	32.9 ± 12.6 (11.9 – 55.3) [b]	1.03 ± 0.40 (0.63 – 1.92) [b]	0.52 (0.02) [c]	4.3 ± 3.4 (0.7 – 11.1) [b]
Guangzhou (n=7) [a]	47.2 ± 5.4 (39.6 – 55.4) [b]	51.9 ± 11.1 (34.2 ± 66.2) [b]	0.91 ± 0.17 (0.79 – 1.24) [b]	0.73 (0.06) [c]	1.2 ± 1.0 (0.5 – 3.1) [b]
Shanghai (n=20) [a]	50.3 ± 17.9 (18.9 – 138.7) [b]	38.2 ± 18.8 (11.4 – 75.3) [b]	1.32 ± 0.43 (0.67 – 2.25) [b]	0.77 (0.00) [c]	1.3 ± 1.1 (0.2 – 4.1) [b]
Xi’an (n=21) [a]	78.7 ± 49.3 (31.0 – 224.9) [b]	109.3 ± 64.4 (49.0 – 305.6) [b]	0.72 ± 0.23 (0.34 – 1.35) [b]	0.89 (0.00) [c]	0.8 ± 0.8 (0.1 – 3.0) [b]

[a]: Number of samples.

[b]: Range of parameter.

[c]: *p*-value.

Table 2. Intercomparison of indoor $PM_{2.5}$ concentrations from different studies.

City/Study Period	No. of Samples	Average Indoor $PM_{2.5}$ Concentration (μg m^{-3})	I/O Ratio	Reference
Hong Kong S.A.R., China				
1999-2000 Winter	34	45.0	0.96	(Chao and Wong, 2002)
2002-2003 Winter	3	73.9	0.94	(Ho et al., 2004)
2016-2017 Winter	20	34.0	1.03	(This Study)
Guangzhou, China				
2003 Summer	9	47.4	1.17	(Lai et al., 2010)
2004-2005 Winter	9	109.9	0.93	(Cao et al., 2012)
2016-2017 Winter	7	47.2	0.91	(This Study)
Shanghai, China				
2013-2014 Winter	47	69.9 [a]	0.83	(Zhou et al., 2018)
2016-2017 Winter	20	50.3	1.32	(This Study)
Xi'an, China				
2007 Winter	3	237.2	0.89	(Zhu et al., 2012)
2016-2017 Winter	21	78.7	0.72	(This Study)

[a] Statistical median value.

Table 3. Summary of indoor OC and EC concentrations and ranges in four cities.

	OC Conc. ($\mu g\ m^{-3}$)	OC Conc. Range ($\mu g\ m^{-3}$)	EC Conc. ($\mu g\ m^{-3}$)	EC Conc. Range ($\mu g\ m^{-3}$)
Hong Kong	12.9 ± 5.1	7.1 – 27.5	3.1 ± 1.7	1.0 – 8.3
Guangzhou	15.8 ± 5.9	7.1 – 21.8	3.6 ± 1.0	2.2 – 4.9
Shanghai	14.6 ± 6.2	5.4 – 29.2	3.7 ± 1.7	1.4 – 6.9
Xi’an	23. 5 ± 9.9	9.0 – 45.9	6.7 ± 4.0	2.3 – 16.7

Table 4. Percentage of OC, EC, and OM in $PM_{2.5}$ mass, OC-EC correlations in indoors and outdoors.

	% OC	% EC	% OM [a]	OC/EC Ratio	OC-EC Correlation (*r*)
Hong Kong					
Indoor	38.5 ± 7.8	8.9 ± 2.3	53.9 ± 10.9	4.53 ± 1.14	0.95 [b]
Outdoor	30.2 ± 9.1	8.2 ± 4.0	42.3 ± 12.7	4.24 ± 1.59	0.68 [c]
Guangzhou					
Indoor	33.9 ± 10.5	7.8 ± 1.2	47.5 ± 14.7	4.31 ± 0.96	0.85 [c]
Outdoor	27.4 ± 5.0	9.9 ± 1.4	38.4 ± 7.0	2.78 ± 0.38	0.94 [c]
Shanghai					
Indoor	29.5 ± 9.2	7.5 ± 3.0	41.3 ± 12.8	4.19 ± 1.15	0.86 [b]
Outdoor	26.9 ± 8.8	7.4 ± 3.9	37.7 ± 12.3	3.96 ± 1.05	0.53
Xi'an					
Indoor	32.4 ± 8.9	8.6 ± 2.7	45.3 ± 12.4	3.91 ± 1.10	0.79 [b]
Outdoor	24.0 ± 7.4	7.1 ± 2.3	33.6 ± 10.4	3.70 ± 1.67	0.86 [b]

[a] OM = OC × 1.4 for urban environment (Chow et al., 2015)

[b] Pearson correlation is significant at the 0.01 level (2-tailed).

[c] Pearson correlation is significant at the 0.05 level (2-tailed).

Table 5. Average indoor and outdoor $PM_{2.5}$ concentrations by three activity classifications.

Indoor Activity	City (Sample Size)	Indoor $PM_{2.5}$ Concentration (μg/m^3)	Outdoor $PM_{2.5}$ Concentration (μg/m^3)	I/O Ratio	Indoor-Outdoor $PM_{2.5}$ Pearson Correlation Coefficient (*r*)	*p*-value of Pearson Correlation
Type A: no specific combustion activity (n=5)	Hong Kong (3)	20.0 ± 10.8	18.9 ± 8.4	1.06	-	-
	Shanghai (2)	31.1 ± 17.3	38.1 ± 37.8	0.82	-	-
	Average	**24.4 ± 13.0**	**26.6 ± 22.4**	**0.92**	-	-
Type B: with only cooking activities (n=39)	Hong Kong (12)	33.4 ± 10.2	36.2 ± 13.4	0.92	0.56	0.06
	Guangzhou (4)	46.2 ± 3.5	51.8 ± 11.9	0.89	0.74	0.26
	Shanghai (10)	42.7 ± 18.0	35.9 ± 18.5	1.19	0.84	0.00
	Xi'an (13)	75.2 ± 42.9	103.7 ± 47.0	0.73	0.94	0.00
	Average	**51.0 ± 31.7**	**60.2 ± 42.8**	**0.85**	-	-
Type C: with cooking and other combustion activities (tobacco smoking or incense burning) (n=22)	Hong Kong (4)	45.8 ± 22.6	35.2 ± 5.4	1.30	0.41	0.59
	Guangzhou (3)	48.5 ± 8.1	52.0 ± 12.5	0.93	-	-
	Shanghai (7)	54.0 ± 19.5	36.3 ± 11.1	1.49	0.86	0.01
	Xi'an (8)	84.4 ± 61.0	118.5 ± 88.9	0.71	0.88	0.00
	Average	**62.8 ± 41.4**	**68.1 ± 65.1**	**0.92**	-	-

Table 6. Average indoor $PM_{2.5}$ associated OC and EC concentrations by three activity classifications.

City	OC Conc. (μg/m^3)	Percentage of OC in $PM_{2.5}$ (%)	EC Conc. (μg/m^3)	Percentage of EC in $PM_{2.5}$ (%)	Percentage of OM [a] in $PM_{2.5}$ (%)	OC/EC Ratio	OC-EC Correlation (r)
Type A							
Hong Kong (2)	10.8 ± 5.2	43.2 ± 5.3	2.7 ± 1.0	10.8 ± 0.1	60.5 ± 7.5	4.02 ± 0.46	-
Shanghai (1)	5.4 ± 0.0	28.6 ± 0.0	1.9 ± 0.0	10.0 ± 0.0	40.0 ± 0.0	2.86 ± 0.00	-
Average	**9.0 ± 4.8**	**39.2 ± 8.7**	**2.4 ± 0.9**	**10.5 ± 0.4**	**54.9 ± 12.2**	**3.72 ± 0.70**	-
Type B							
Hong Kong (10)	11.2 ± 3.0	33.4 ± 8.4	2.4 ± 0.9	7.2 ± 2.0	46.8 ± 11.7	4.64 ± 1.27	0.81 [b]
Guangzhou (4)	15.4 ± 5.1	33.3 ± 10.1	3.5 ± 0.9	7.5 ± 1.6	46.6 ± 14.1	4.43 ± 0.91	-
Shanghai (7)	14.9 ± 6.0	30.2 ± 10.3	4.1 ± 1.9	8.3 ± 2.8	42.3 ± 14.5	3.66 ± 1.21	0.88 [b]
Xi'an (11)	17.9 ± 7.3	21.6 ± 2.9	4.5 ± 1.6	5.4 ± 1.4	30.2 ± 4.0	4.00 ± 0.62	0.94 [b]
Average	**16.2 ± 7.8**	**29.1 ± 8.4**	**4.1 ± 2.4**	**7.4 ± 2.1**	**40.8 ± 11.8**	**3.96 ± 1.08**	**0.94 [b]**
Type C							
Hong Kong (4)	18.0 ± 7.2	39.3 ± 6.0	5.2 ± 2.5	11.1 ± 2.1	55.0 ± 8.4	3.54 ± 0.44	-
Guangzhou (3)	16.4 ± 8.1	33.8 ± 13.1	3.8 ± 1.2	7.8 ± 1.4	47.3 ± 18.3	4.31 ± 1.18	-
Shanghai (7)	13.6 ± 2.8	25.1 ± 7.3	3.3 ± 1.3	6.1 ± 2.4	35.1 ± 10.3	4.13 ± 1.16	0.85 [c]
Xi'an (8)	25.8 ± 10.4	30.5 ± 10.0	7.8 ± 5.1	9.3 ± 3.3	42.7 ± 14.1	3.30 ± 1.62	0.71 [c]
Average	**19.2 ± 9.0**	**30.5 ± 9.8**	**5.3 ± 3.8**	**8.5 ± 3.1**	**42.8 ± 13.8**	**3.60 ± 1.23**	**0.81 [b]**

[a] OM = OC × 1.4 for urban environment (Chow et al., 2015)

[b] Pearson correlation is significant at the 0.01 level (2-tailed).

[c] Pearson correlation is significant at the 0.05 level (2-tailed).

Note: Due to the limited sample size, all Type A, Type B household in Guangzhou, and Type C household in Hong Kong and Guangzhou were excluded for the correlation comparison.

References

Cao, J.J., Huang, H., Lee, S.C., Chow, J.C., Zou, C.W., Ho, K.F., Watson, J.G. (2012). Indoor/Outdoor Relationships for Organic and Elemental Carbon in $PM_{2.5}$ at Residential Homes in Guangzhou, China. Aerosol Air Qual. Res. 12, 902-910. https://doi.org/10.4209/aaqr.2012.02.0026.

Chao, C.Y., Wong, K.K. (2002). Residential indoor PM_{10} and $PM_{2.5}$ in Hong Kong and the elemental composition. Atmos. Environ. 36, 265-277. https://doi.org/10.1016/S1352-2310(01)00411-3.

Chow, J.C., Lowenthal, D.H., Chen, L.-W.A., Wang, X., Watson, J.G. (2015). Mass reconstruction methods for $PM_{2.5}$: a review. Air Qual. Atmos. Health 8, 243-263. https://doi.org/10.1007/s11869-015-0338-3.

Ho, K.F., Cao, J.J., Harrison, R.M., Lee, S.C., Bau, K.K. (2004). Indoor/outdoor relationships of organic carbon (OC) and elemental carbon (EC) in $PM_{2.5}$ in roadside environment of Hong Kong. Atmos. Environ. 38, 6327-6335. https://doi.org/10.1016/j.atmosenv.2004.08.007.

Lai, S.C., Ho, K.F., Zhang, Y.Y., Lee, S.C., Huang, Y., Zou, S.C. (2010). Characteristics of Residential Indoor Carbonaceous Aerosols: A Case Study in Guangzhou, Pearl River Delta Region. Aerosol Air Qual. Res. 10, 472-478. https://doi.org/10.4209/aaqr.2010.05.0037.

Zhou, X., Cai, J., Zhao, Y., Chen, R., Wang, C., Zhao, A., Yang, C., Li, H., Liu, S., Cao, J., Kan, H., Xu, H. (2018). Estimation of residential fine particulate matter infiltration in Shanghai, China. Environ. Pollut. 233, 494-500. https://doi.org/10.1016/j.envpol.2017.10.054.

Zhu, C.-S., Cao, J.-J., Shen, Z.-X., Liu, S.-X., Zhang, T., Zhao, Z.-Z., Xu, H.-M., Zhang, E.-K. (2012). Indoor and Outdoor Chemical Components of $PM_{2.5}$ in the Rural Areas of Northwestern China. Aerosol Air Qual. Res. 12, 1157-1165. https://doi.org/10.4209/aaqr.2012.01.0003.

Figure Captions

Fig. 1. Locations of investigated four cities.

Fig. 2. (a) Indoor and outdoor $PM_{2.5}$ concentration and $PM_{2.5}$ I/O ratio, and (b) relation between outdoor $PM_{2.5}$ concentration and I/O ratio during periods of different degrees of pollution.

Fig. 3. Portions of OC and EC in indoor $PM_{2.5}$ mass during periods of different degrees of pollution.

Fig. 4. Temporal variations of indoor $PM_{2.5}$ concentrations for (a) Type A household in Shanghai; (b) Type B household in Xi'an; and (c) Type C household in Hong Kong, Missing data for Type B (XA-06) and Type C (HK-05) were due to pausing of measurement during filter changing.

Fig. 5. Contributions of indoor activities to indoor $PM_{2.5}$ concentrations of residential homes in Hong Kong and Shanghai.

Fig. 1. Locations of investigated four cities.

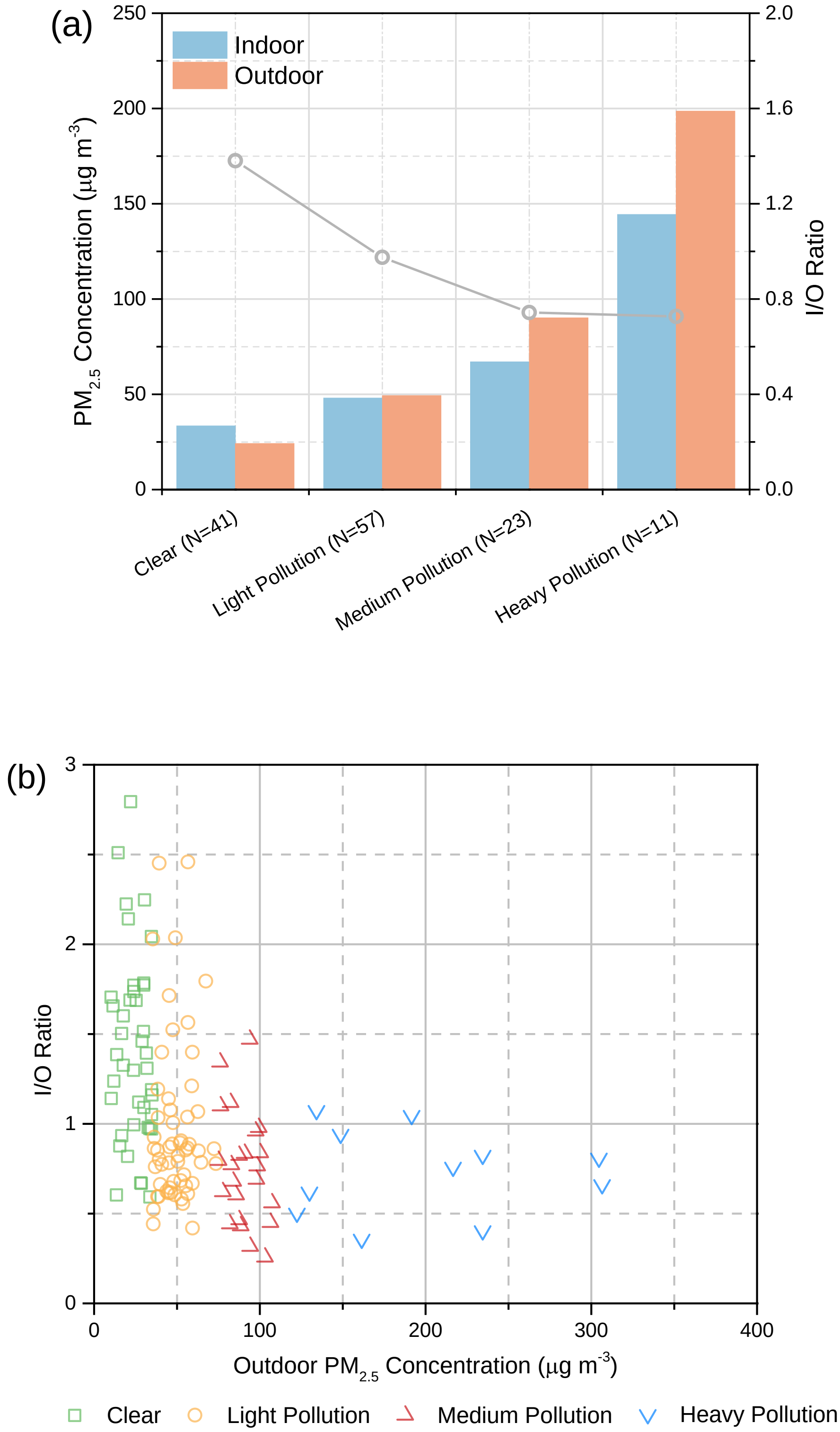


Fig. 2. (a) Indoor and outdoor $PM_{2.5}$ concentration and $PM_{2.5}$ I/O ratio, and (b) relation between outdoor $PM_{2.5}$ concentration and I/O ratio during periods of different degrees of pollution.

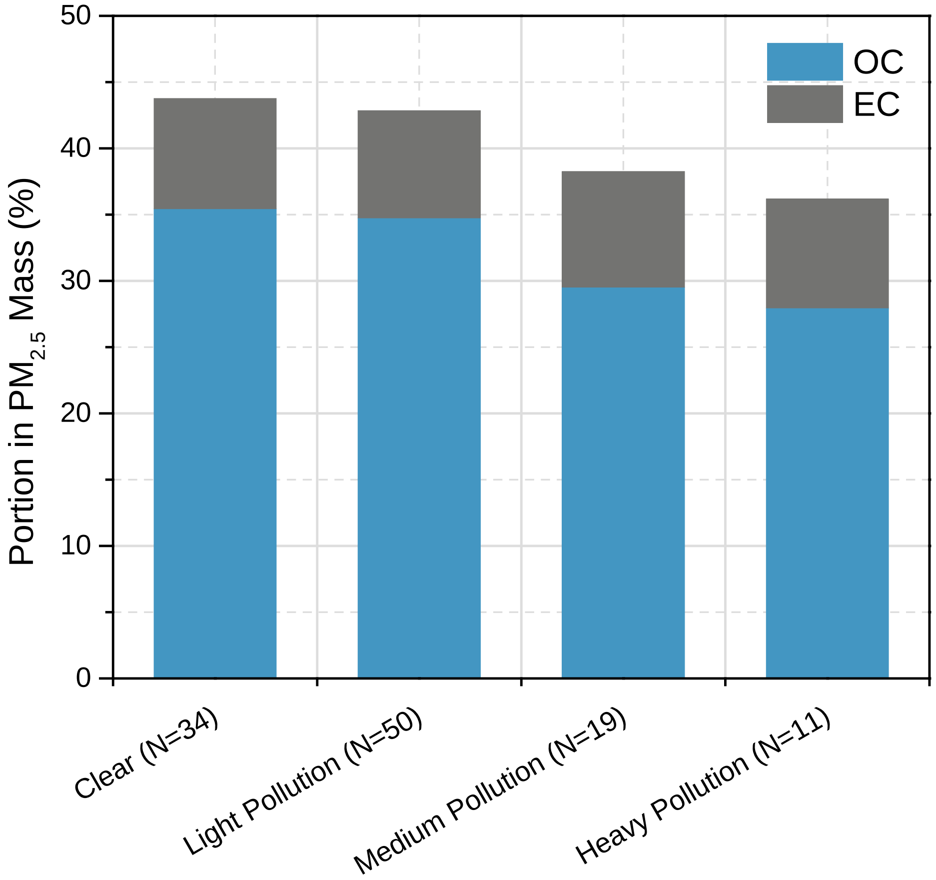


Fig. 3. Portions of OC and EC in indoor $PM_{2.5}$ mass during periods of different degrees of pollution.

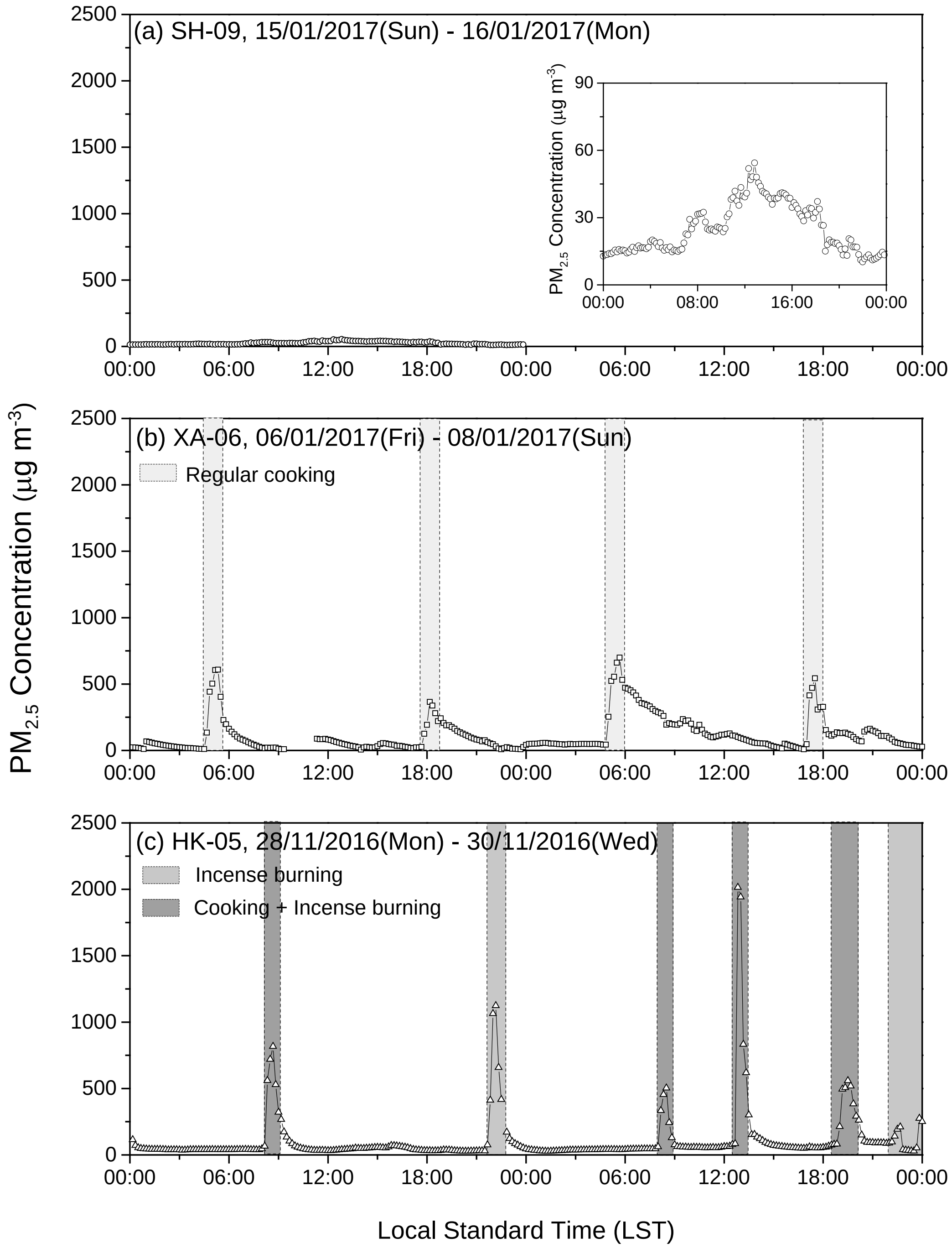


Fig. 4. Temporal variations of indoor $PM_{2.5}$ concentrations for (a) Type A household in Shanghai; (b) Type B household in Xi'an; and (c) Type C household in Hong Kong, Missing data for Type B (XA-06) and Type C (HK-05) were due to pausing of measurement during filter changing.

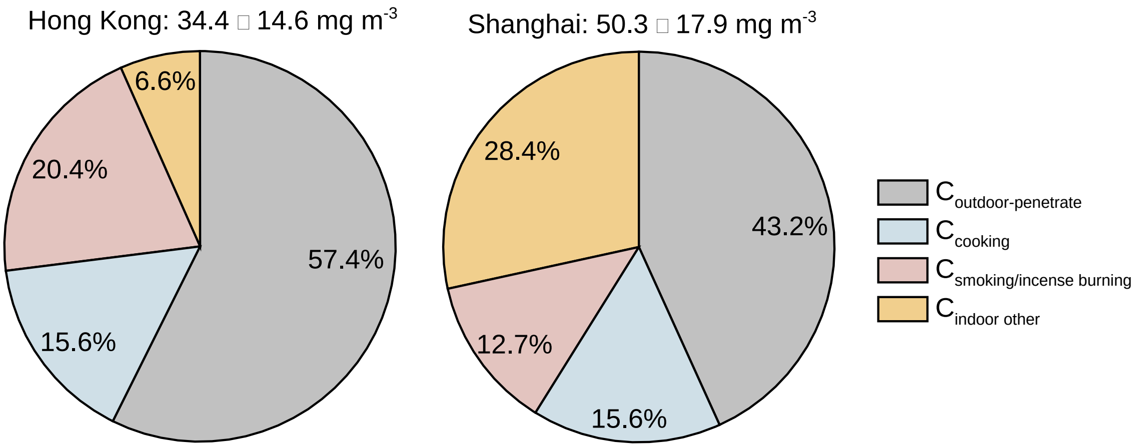


Fig. 5. Contributions of indoor activities to indoor $PM_{2.5}$ concentrations of residential homes in Hong Kong and Shanghai.

Effects of Indoor Activities and Outdoor Penetration on $PM_{2.5}$ and Associated Organic/Elemental Carbon at Residential Homes in Four Chinese Cities during Winter

Zhuo-zhi Zhang[1], Yuan Gao[1,2], Qi Yuan[1], Yan Tan[1], Hai-wei Li[1,3], Long Cui[4], Yu Huang[4], Guang-li Xiu[5], Sen-chao Lai[6], Judith C. Chow[7], John G. Watson[7], Shun-cheng Lee[1]*

[1] Department of Civil and Environmental Engineering, The Hong Kong Polytechnic University, Hung Hom, Hong Kong

[2] Department of Civil Engineering, Chu Hai College of Higher Education, Tun Mun, Hong Kong

[3] Jiangsu Key Laboratory of Atmospheric Environment Monitoring and Pollution Control (AEMPC), Collaborative Innovation Center of Atmospheric Environment and Equipment Technology (CIC-AEET), School of Environmental Science and Engineering, Nanjing University of Information Science & Technology, Nanjing, 210044, China

[4] Key Lab of Aerosol Chemistry & Physics, SKLLQG, Institute of Earth Environment, Chinese Academy of Sciences, Xi'an, China

[5] School of Resources and Environmental Engineering, East China University of Science and Technology, Shanghai, China

[6] School of Environment and Energy, South China University of Technology, Guangzhou, China

[7] Division of Atmospheric Sciences, Desert Research Institute, Reno, NV, USA

Submitted to Science of the Total Environment

* Corresponding Author

Shun-cheng Lee, Ph.D.

Tel: +852-2766-6011

Email: shun-cheng.lee@polyu.edu.hk

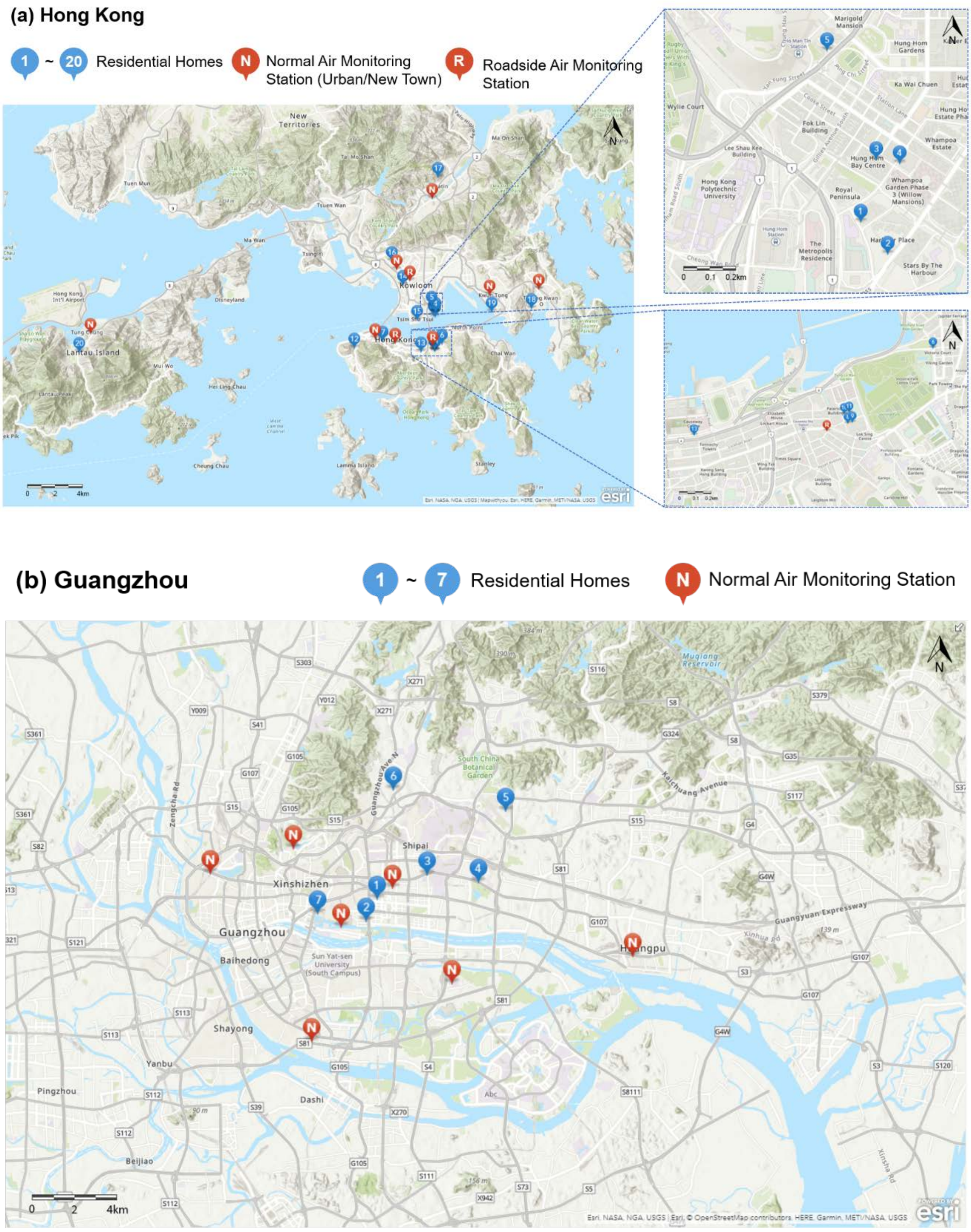
Table S1. Characteristics of Residential Homes in Four Cities

City	Home ID	Type [a]	Site Description	Location	Elevation	Floor Area (m^2)	Number of Residents	Fuel Type	Incense Burning	Smoker	Indoor/ Outdoor Temperature
Hong Kong	HK-01	A	Near road with medium traffic flow	Urban	19/F	10	2	No	No	No	22.0/19.0
	HK-02	B	Near road with low traffic flow	Urban	7/F	60	6	LPG	No	No	21.6/25.4
	HK-03	A	Near road with medium traffic flow	Urban	2/F	30	6	No	No	No	22.0/25.5
	HK-04	A	Near road with medium traffic flow, restaurant nearby	Urban	3/F	13	2	No	No	No	22.0/20.3
	HK-05	C	Near main road with heavy traffic flow	Urban	6/F	30	2	LPG	Yes	No	21.6/19.1
	HK-06	B	Near main road with heavy traffic flow	Urban	15/F	100	4	LPG	No	1[b]	21.8/21.2
	HK-07	B	Near road with low traffic flow	Urban	18/F	70	4	LPG	No	No	22.1/19.7
	HK-08	C	Near road with medium traffic flow	Urban	10/F	45	3	Electricity	No	1[c]	22.5/20.9
	HK-09	B	Near road with medium traffic flow	Urban	16/F	53	2	LPG	No	No	21.5/18.8
	HK-10	B	Near road with medium traffic flow	Urban	23/F	35	4	LPG	No	No	23.4/20.2
	HK-11	C	Near road with medium traffic flow	Urban	26/F	50	2	LPG	No	1[c]	20.0/17.2
	HK-12	N/A	Near road with low traffic flow	Urban	2/F	50	3	No	No	1[c]	24.4/20.9
	HK-13	B	Near main road with heavy traffic flow	Urban	15/F	30	1	LPG	No	No	24.0/19.6
	HK-14	B	Near road with low traffic flow	Urban	15/F	70	3	LPG	No	No	21.6/15.7
	HK-15	C	Near main road with heavy traffic flow	Urban	4/F	70	3	LPG	No	3[c]	26.4/18.4
	HK-16	B	Near road with medium traffic flow	Urban	15/F	70	3	LPG	No	No	19.1/13.4
	HK-17	B	Residential area, far from the main road	Suburban	6/F	150	4	LPG	No	No	20.9/18.1
	HK-18	B	Residential area, far from the main road	Suburban	18/F	65	3	LPG	No	No	21.7/18.5
	HK-19	B	Residential area, far from the main road	Urban	25/F	50	2	LPG	No	No	22.1/18.6
	HK-20	B	Residential area, far from the main road	Suburban	22/F	50	5	LPG	No	No	21.7/16.9
Guangzhou	GZ-01	B	Residential area, far from the main road	Urban	13/F	120	4	NG	No	No	19.5/16.6
	GZ-02	C	Near road with low traffic flow	Urban	21/F	75	3	LPG	No	1[c]	23.8/19.0
	GZ-03	C	Near road with low traffic flow	Urban	10/F	97	3	NG	Yes	1[c]	16.8/15.3
	GZ-04	B	Near road with low traffic flow	Suburban	28/F	120	3	LPG	No	No	22.3/16.1
	GZ-05	B	Near main road with heavy traffic flow	Suburban	4/F	130	3	NG	No	1[b]	16.8/15.4
	GZ-06	B	Near road with low traffic flow	Suburban	5/F	130	3	LPG	No	No	23.8/20.2
	GZ-07	C	Near main road with heavy traffic flow	Urban	28/F	50	3	LPG	Yes	No	22.1/17.2
Shanghai	SH-01	B	Residential area, far from the main road	Suburban	6/F	90	4	LPG	No	No	N/A/9.8
	SH-02	C	Residential area, far from the main road	Suburban	1/F	90	7	LPG	No	2[c]	N/A/9.2
	SH-03	B	Near road with medium traffic flow	Suburban	9/F	80	3	LPG	No	No	15.9/11.8
	SH-04	B	Residential area, far from the main road (industrial)	Suburban	12/F	50	4	LPG	No	No	17.8/12.6
	SH-05	N/A	Residential area, far from the main road (industrial)	Suburban	17/F	50	1	No	No	1[c]	13.1/8.1

Shanghai	SH-06	A	Residential area, far from the main road (industrial)	Suburban	17/F	50	1	No	No	1[c]	17.4/9.0
	SH-07	B	Near road with medium traffic flow	Suburban	20/F	50	1	No	No	No	17.1/10.1
	SH-08	B	Near road with light traffic flow	Suburban	3/F	90	3	LPG	No	No	12.7/6.0
	SH-09	A	Near road with light traffic flow	Suburban	5/F	120	3	LPG	No	No	8.8/6.3
	SH-10	B	Residential area, far from the main road	Urban	4/F	120	3	No	No	No	11.6/10.2
	SH-11	C	Residential area, far from the main road	Suburban	8/F	90	3	LPG	No	No	15.5/9.1
	SH-12	B	Residential area, far from the main road	Suburban	2/F	70	3	LPG	No	1[c]	11.3/5.2
	SH-13	B	Near road with medium traffic flow (industrial)	Suburban	2/F	90	3	LPG	No	No	14.4/8.7
	SH-14	C	Near road with medium traffic flow (industrial)	Suburban	2/F	180	2	LPG	No	No	15.0/8.0
	SH-15	B	Near main road with low traffic flow	Rural	5/F	70	3	LPG	No	1[c]	13.1/8.8
	SH-16	C	Near main road with low traffic flow	Rural	1/F	220	3	LPG	No	1[b]	13.0/10.6
	SH-17	B	Near main road with low traffic flow	Rural	2/F	200	3	LPG	No	1[c]	11.8/11.0
	SH-18	C	Near main road with low medium flow	Rural	1/F	220	5	LPG	No	No	13.8/12.2
	SH-19	C	Near main road with low traffic flow	Rural	1/F	190	1	LPG	No	2[c]	17.9/13.1
	SH-20	C	Near main road with medium traffic flow	Urban	1/F	220	3	LPG	No	1[c]	18.4/20.1
Xi’an	XA-01	B	Near road with medium traffic flow	Suburban	1/F	120	5	NG	No	No	24.9/10.5
	XA-02	C	Near road with heavy traffic flow	Suburban	26/F	80	2	LPG	No	1[c]	12.3/2.7
	XA-03	B	Residential area with low traffic flow	Suburban	6/F	100	3	NG	No	No	20.3/7.4
	XA-04	B	Residential area with low traffic flow	Suburban	18/F	76	3	LPG	No	No	19.1/4.0
	XA-05	C	Near road with heavy traffic flow (industrial)	Suburban	23/F	80	5	NG	No	1[c]	20.1/3.1
	XA-06	B	Near road with medium traffic flow	Suburban	4/F	100	4	Electricity	No	No	14.3/3.3
	XA-07	B	Near road with heavy traffic flow	Urban	23/F	110	2	LPG	No	No	18.7/0.7
	XA-08	C	Residential area with low traffic flow	Suburban	5/F	50	3	LPG	No	1[c]	17.6/1.9
	XA-09	C	Residential area with low traffic flow (industrial)	Suburban	24/F	130	3	NG	No	1[c]	19.1/1.1
	XA-10	B	Near road with medium traffic flow	Urban	5/F	60	2	NG	No	No	19.1/9.8
	XA-11	B	Near road with heavy traffic flow (industrial)	Suburban	4/F	60	2	LPG	No	No	15.6/10.8
	XA-12	B	Near road with heavy traffic flow	Suburban	2/F	90	2	NG	No	No	14.3/4.4
	XA-13	C	Near road with heavy traffic flow	Suburban	3/F	100	2	LPG	No	1[c]	21.7/6.4
	XA-14	C	Near road with medium traffic flow (industrial)	Suburban	8/F	90	3	LPG	No	1[c]	20.5/8.5
	XA-15	B	Near road with medium traffic flow (industrial)	Suburban	6/F	90	3	LPG	No	No	17.6/7.3
	XA-16	B	Residential area with low traffic flow	Suburban	2/F	80	2	LPG	No	No	16.1/10.1
	XA-17	B	Near road with heavy traffic flow	Urban	5/F	80	4	NG	No	No	20.1/9.2
	XA-18	C	Residential area with low traffic flow (industrial)	Suburban	5/F	120	3	NG	No	2[c]	20.1/5.5
	XA-19	C	Near road with medium traffic flow (industrial)	Suburban	8/F	110	2	NG	No	1[c]	23.7/9.4
	XA-20	B	Residential area with low traffic flow (industrial)	Suburban	1/F	90	2	NG	No	No	18.9/9.8
	XA-21	B	Residential area with low traffic flow (industrial)	Suburban	4/F	150	2	NG	No	No	19.8/10.3

Note:

LPG: Liquified Petroleum Gas
NG: Natural Gas
[a]: **A** for no specific combustion activities, **B** for with only cooking activities, **C** for with cooking and other indoor combustion activities such as tobacco smoking and incense burning
[b]: Smoker smoking outside the room
[c]: Smoker smoking inside the room


(a) Hong Kong
1 ~ 20 Residential Homes
N Normal Air Monitoring Station (Urban/New Town)
R Roadside Air Monitoring Station
New Territories
Tuen Mun
Tsuen Wan
Ma Wan
Tsing Yi
Hong Kong Int'l Airport
Tung Chung
Lantau Island
Disneyland
Kowloon
Tsim Sha Tsui
Kwun Tong
Hong Kong
Chai Wan
Cheung Chau
Lamma Island
Stanley
Ma On Shan
Mui Wo
Marigold Mansion
Hung Hom Gardens
Ka Wai Chuen
Wylie Court
Fok Lin Building
Lee Shau Kee Building
Hong Kong Polytechnic University
Hung Hom Bay Centre
Whampoa Estate
Whampoa Garden Phase 3 (Willow Mansions)
Royal Peninsula
Hung Hom Station
The Metropolis Residence
Stars By The Harbour
0 2 4km
0 0.1 0.2km
(b) Guangzhou
1 ~ 7 Residential Homes
N Normal Air Monitoring Station
South China Botanical Garden
Shipai
Xinshizhen
Guangzhou
Baihedong
Sun Yat-sen University (South Campus)
Shayong
Yanbu
Pingzhou
Dashi
Beijiao
Huangpu
Guangyuan Expressway
Kaichuang Avenue
Zengcha Rd
Mugiang Reservoir
0 2 4km
Esri, NASA, NGA, USGS | Esri, © OpenStreetMap contributors, HERE, Garmin, METI/NASA, USGS

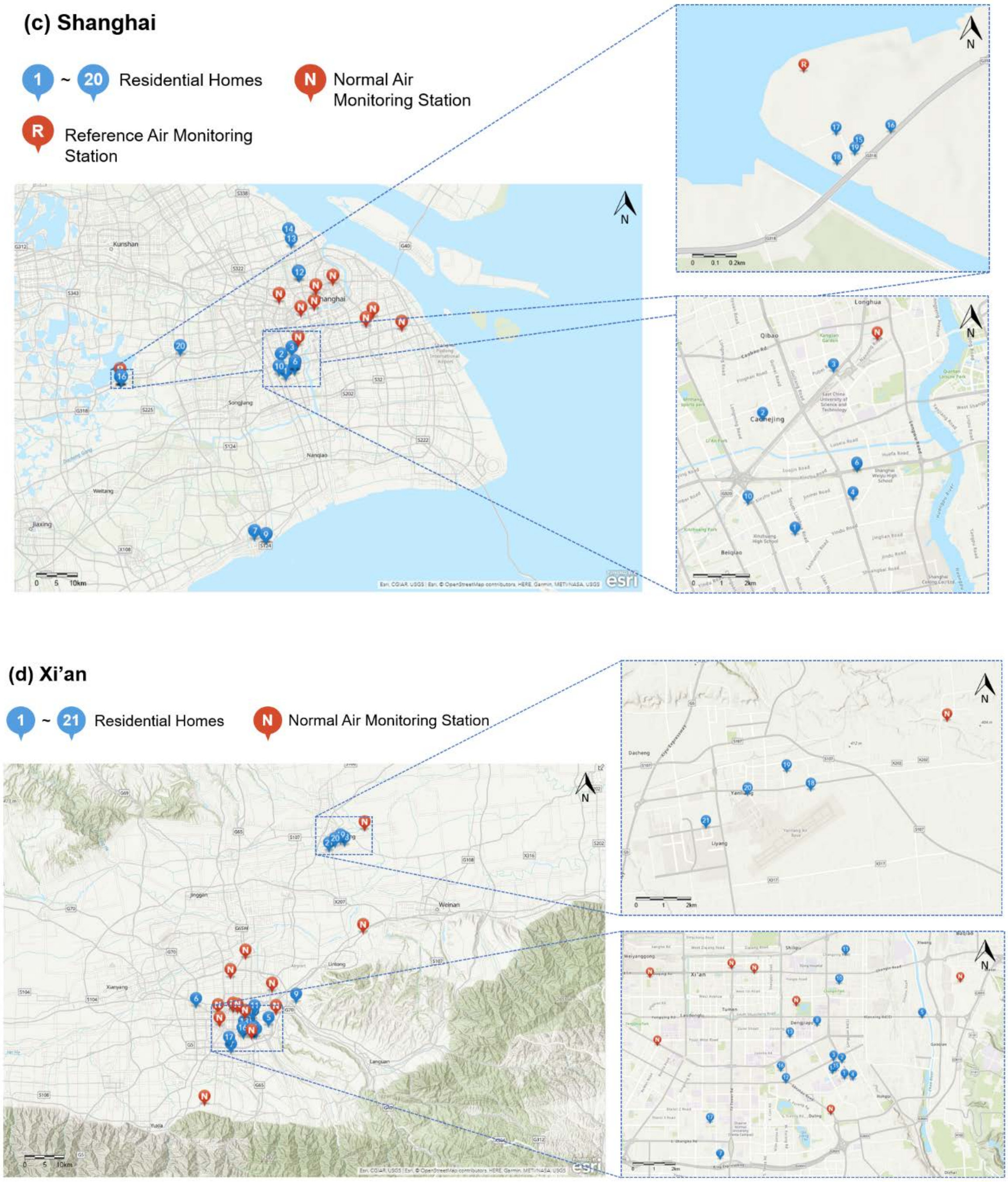


Fig. S1. Locations of residential homes and nearby air quality monitoring stations in (a) Hong Kong, (b) Guangzhou, (c) Shanghai and, (d) Xi’an.