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Determinants of personal exposure to fine particulate matter ($PM_{2.5}$) in adult subjects in Hong Kong

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Abstract

Personal monitoring for fine particulate matter ($PM_{2.5}$) was conducted for adults (48 subjects, 18-63 years of age) in Hong Kong during the summer and winter of 2014-2015. All filters were analyzed for $PM_{2.5}$ mass and constituents (including carbonaceous aerosols, water-soluble ions, and elements). We found that season (p = 0.02) and occupation (p < 0.001) were significant factors affecting the strength of the personal-ambient $PM_{2.5}$ associations. We applied mixed-effects models to investigate the determinants of personal exposure to $PM_{2.5}$ mass and constituents, along with within- and between-individual variance components. Ambient $PM_{2.5}$ was the dominant predictor of (R^2 = 0.12-0.59, p < 0.01) and the largest contributor (> 37.3%) to personal exposures for $PM_{2.5}$ mass and most components. For all subjects, a one-unit (2.72 ug/m^3) increase in ambient $PM_{2.5}$ was associated with a 0.75 ug/m^3 (95% CI: 0.59-0.94 ug/m^3) increase in personal $PM_{2.5}$ exposure. The adjusted mixed-effects models included information extracted from individual's activity diaries as covariates. The results showed that season, occupation, time indoors at home, in transit, and cleaning were significant determinants for $PM_{2.5}$ components in personal exposure (R^2_β = 0.06-0.63, p < 0.05), contributing to 3.0-70.4% of the variability. For one-hour extra time spent at home, in transit, and cleaning an average increase of 1.7-3.6% (ammonium, sulfate, nitrate, sulfur), 2.7-12.3% (elemental carbon, ammonium, titanium, iron), and 8.7-19.4% (ammonium, magnesium ions, vanadium) in components of personal $PM_{2.5}$ were observed, respectively. In this research, the within-individual variance component dominated the total variability for all investigated exposure data except $PM_{2.5}$ and EC. Results from this study indicate that performing long-term personal monitoring is needed for examining the associations of mass and constituents of personal $PM_{2.5}$ with health outcomes in

epidemiological studies by describing the impacts of individual-specific data on personal exposures.

Keywords: personal exposure; fine particulate matter; particulate constituents; time-activity diaries; mixed-effects model

1. Introduction

Previous epidemiological studies in Europe and North America have revealed that ambient concentration of fine particles ($PM_{2.5}$, with aerodynamic diameters less than 2.5 μm) and chemical components in $PM_{2.5}$, including elemental and organic carbon, sulfate, nitrate, and trace elements, showed significant correlations with adverse health effects (Franklin et al., 2006; Kim et al., 2015; Pope et al., 2002; Rohr and Wyzga, 2012; WHO, 2013). Similar conclusions have been obtained in Hong Kong and other Chinese cities, such as links between increased hospitalization and mortality from respiratory diseases with high particulate matter (PM_{10}/$PM_{2.5}$) pollution levels (Cao et al., 2012; Pun et al., 2014; Xie et al., 2016). Human exposure depends on the amount of time an individual spends in indoor microenvironments, outdoors, transit, in addition to personal activities (e.g., time spent cooking and cleaning, proximity to local sources that cannot be captured by the general monitoring sites) (Jiao et al., 2012; Ott et al., 2010; Wallace et al., 2006). Consequently, using stationary ambient concentration as a proxy for personal exposure has raised concerns. Because it may lead to potential misclassification of total personal exposures (Avery et al., 2010b; Hsu et al., 2012; Wilson and Brauer, 2006), and bias the exposure-response relationship in epidemiological studies (Ji and Zhao, 2015; Meng et al., 2005).

Previous exposure studies have focused on assessing $PM_{2.5}$/PM_{10} in personal exposures and residential indoor/outdoor (Clayton et al., 1993; Johannesson et al., 2007; Williams et al., 2000;

Xu et al., 2014). Some of these studies have also measured personal exposure to $PM_{2.5}$ components, such as sulfate, elemental carbon, and trace elements from the susceptible populations (Du et al., 2010; Janssen et al., 2005; Noullett et al., 2006) and healthy adults (Chen et al., 2017a; Du et al., 2010; Montagne et al., 2014). Several studies have examined the personal-ambient correlations, which exhibited a large spread between different studies, but overall relationships were stronger for longitudinal studies (Adgate et al., 2003; Jahn et al., 2013; Kim et al., 2005a; Suh and Zanobetti, 2010) compared to cross-sectional studies (Avery et al., 2010a; Janssen et al., 2005). A few studies have also characterized the factors influencing the strength of associations between ambient concentrations and corresponding personal exposures (Brown et al., 2008; Meng et al., 2009; Ozkaynak et al., 1996).

Personal exposures can vary widely, for the same ambient concentration, across individuals in a given community or city and within individuals over time (Jahn et al., 2013; Tunno et al., 2016). Insufficient attention to the balance of within-individual (σ^2_w) and between-individual variance (σ^2_b) in personal exposure can reduce the efficiency of measurement efforts and attenuate estimates of exposure-response associations (Loomis and Kromhout, 2004). Thus, it is essential to obtain repeated personal measurements from study subjects to accurately estimate exposure-response relationships, especially in epidemiological studies (Baccarelli et al., 2014; Lanki et al., 2007; Nieuwenhuijsen, 2015). For example, Johannesson et al. (2011) have characterized the degree of variability in σ^2_w and σ^2_b to estimate the number of repeated personal measurements per participant that would need to restrict the attenuation bias to 20% among a Swedish population. Moreover, intra-class correlation coefficient (ICC), which represents the proportion of the total variance attributed to between-individual variation, has been discussed to quantify the accuracy of measurements (Xu et al., 2016). Questionnaires and activity diaries have

been used to collect information on factors influencing between- and within-individual variance in exposure assessment studies (Johannesson et al., 2011; Lanki et al., 2007; Scapellato et al., 2009). These issues are important in consideration of the study design and interpretation of personal monitoring to assess general population levels (e.g., individuals with high or low exposures) and to investigate possible associations between personal exposures and health risks.

Past studies investigated the determinants (or factors) affecting personal exposure to $PM_{2.5}$ in susceptible populations, such as elderly, individuals with the cardiovascular or respiratory disease, or children with asthma (Brown et al., 2009; Lanki et al., 2007; Scapellato et al., 2009). These factors, however, are poorly quantified, particularly for $PM_{2.5}$ components in personal exposures among the adult population (Adgate et al., 2007; Johannesson et al., 2011; Sørensen et al., 2005). The determinants of personal exposure to $PM_{2.5}$ mass and components in addition to within- and between-individual variance requires further elucidation. A thorough understanding of the variability and determinants of personal exposure to particulate matter pollution can improve the study design and help in developing targeted risk-reduction strategies in epidemiological studies.

The objectives of this study are to 1) characterize the seasonal and occupational variations of personal exposure to $PM_{2.5}$ mass and components among adult subjects in Hong Kong; 2) assess the factors influencing associations of personal-ambient $PM_{2.5}$; 3) investigate the determinants of personal exposure to $PM_{2.5}$ mass and constituents, as well as to estimate the between- and within-individual variance components using mixed-effects models.

2. Methods

2.1 Study population

Forty-eight (48) adults (18-63 years of age) living and working in different districts of Hong Kong participated in the personal monitoring campaign between July 2014 and March 2015. Advertisements (e.g., via University Mass Mails) and flyers were used to recruit potential participants; the target study subjects were healthy non-smoking adults (> 18 years of age), living in non-smoking homes, residence in Hong Kong for the past twelve months and free from chronic diseases. Forty-two (42) and 41 participants were monitored in summer and winter, respectively, with 73% of the 48 individuals participating in both seasons. The Joint Chinese University of Hong Kong-New Territories East Cluster Clinical Research Ethics Committee approved this study before subject recruitment. Subjects in this study signed informed consent before their participation in the personal monitoring program.

2.2 Personal monitoring and exposure assessment

Personal exposure to $PM_{2.5}$ was measured using a Personal Environmental Monitor (PEM, Model 200, MSP Corp., Shoreview, MN, USA) together with a Leland Legacy pump (SKC Inc., Eighty-Four, PA, USA) and operated at a flow rate of 10 L/min for twenty-four-hour (24-hr) (00:00-24:00, local time). Two PEMs loaded with one Teflon and one quartz filter (37 mm, 2 μm pore size, Pall Corporation, MI, USA), respectively, were carried simultaneously by each subject. PEMs were kept near the breathing zone of the participant to mimic actual personal exposures. Participants were instructed to bring the sampling device with them at all times but were allowed to place the sampler nearby when subjects were at home or work. All study subjects were encouraged to maintain their regular activity patterns during the daily sampling period. Personal monitoring from each subject was conducted in a two-day (e.g., workday, weekend) sampling event within 1-2 weeks intervals from July to October 2014 and December

2014 to March 2015, respectively. This analysis included 48 participants with 2-4 observations from each subject. Altogether, 161 sampling periods (on 102 different days) resulted in a total of 322 filter samples.

Prior to personal sampling, participants were asked to complete a detailed questionnaire regarding personal information such as gender, occupation, and residential characteristics. During the 24-hr monitoring events, each participant was required to fill out a time-activity diary denoting their locations and activities every 15 minutes; research assistant would check the activity dairy after each sampling session. Survey data and activity pattern provided additional information for use in mixed-effects modelling. Time spent indoors (e.g., at home), outdoors, in transit (e.g., on the bus/minibus, in Metro), as well as the amount of time spent cooking and cleaning within their residence, were included in the mixed-effects model as covariates.

Personal exposure to $PM_{2.5}$ mass was determined by gravimetric analyses using a microbalance (Model MC 5-0CE, Sartorius AG, Goettingen, Germany) in a temperature (20-25°C) and humidity ($35 \pm 5\%$) controlled weighing room. Information about sampling performance can be found in Figure S1 (see Supporting Information, SI).

Ambient data were retrieved from the Hong Kong Environmental Protection Department (HKEPD) Air Quality Monitoring Network (http://epic.epd.gov.hk/EPICDI/air/station/), which provides integrated 24-hr $PM_{2.5}$ concentrations from the HKEPD Air Quality Monitoring Stations. Figure S2 shows the location of eleven general air quality monitoring stations (including Central/Western, Eastern, Kwai Chung, Kwun Tong, Sham Shui Po, Tsuen Wan, Sha Tin, Tai Po, Tuen Mun, Tung Chung, Yuen Long) in different districts of Hong Kong. The corresponding distance of ambient monitoring stations and participants' residences ranging from 10.0 to 23.2 km with an average of 13.9 km. It is assumed that these distances (< 20 km) would not affect the

estimated associations (Sarnat et al., 2010). Table S1 of SI summarizes the Spearman's correlations for $PM_{2.5}$ between eleven ambient sites (r_s: 0.78-0.95, $p < 0.01$). Also, Table S2 provides coefficients of divergence across these sites (COD, ranging from 0.01 to 0.29 and 0.02 to 0.19 in summer and winter, respectively). In the present study, cross-sectional means (i.e., 24-hr average ambient $PM_{2.5}$ data across all these sites on the same day) were compared with personal $PM_{2.5}$ exposures.

2.3 Chemical analysis

Organic carbon (OC) and elemental carbon (EC) were analyzed using a DRI Model 2001 Thermal/Optical Carbon Analyzer (Atmoslytic Inc., Calabasas, CA, USA) by thermal/optical reflectance (TOR) following the IMPROVE_A protocol (Chow et al., 2011). The method detection limit (MDL) of OC and EC were 0.28 and 0.04 μg/m^3 respectively. Procedural blank values were subtracted from sample concentrations.

Water-soluble inorganic ions including four anions (chloride (Cl^-), nitrate (NO_3^-), sulfate (SO_4^{2-}), and oxalate ($C_2O_4^{2-}$)) and five cations (sodium (Na^+), ammonium (NH_4^+), potassium (K^+), magnesium (Mg^{2+}), and calcium (Ca^{2+})) were analyzed using a Dionex ICS-3000 Ion Chromatograph (Ho et al., 2014). Average field blanks were subtracted from each sample filter. MDLs of ions were within the range of 0.01 to 0.23 μg/m^3.

A total of 19 elements (including sodium (Na), magnesium (Mg), aluminium (Al), silicon (Si), sulfur (S), chlorine (Cl), potassium (K), calcium (Ca), titanium (Ti), vanadium (V), chromium (Cr), manganese (Mn), iron (Fe), nickel (Ni), copper (Cu), zinc (Zn), arsenic (As), bromine (Br), and lead (Pb)) were analyzed using an Energy Dispersive X-Ray Fluorescence analyzer (ED-XRF, Epsilon 5, PANalytical Company, Netherlands) from Teflon filters following the

gravimetric analyses (Chow and Watson, 2012). The analyses were conducted according to the standard operating procedures at the Desert Research Institute laboratories (DRI, Reno, NV, USA) including quality assurance and quality control (Watson et al., 1999). MDLs of the elements were within the range of 0.5-33 ng/m^3. Although personal $PM_{2.5}$ and components concentrations were the primary analyses, further plans include examining the sources of personal $PM_{2.5}$.

2.4 Quality Assurance/Quality Control

Before the personal monitoring program, quartz filters were baked at 900˚C for 3 h to remove any carbon residue. Triplicate filter weights (within ± 3 μg agreement) were determined after conditioning the filters in a dry box (RH < 40%) for 24-hr before and after sampling. Sampling pumps were calibrated to 10 (± 0.5) L/min before monitoring and measured after sampling using a DryCal DC-Lite flow meter (BIOS Inc., Bulter, NJ, USA). Field blanks were collected without switching on the sampling pump to account for artifacts and contamination during sample collection, and seven sets of Teflon and quartz filter blanks were collected during the summer and winter campaigns, respectively. All filter samples and blanks were labeled immediately and stored in a desiccator before sampling. After sample collection, all filters were stored in a freezer (-20 ˚C) to minimize semi-volatile losses.

2.5 Statistical analysis

Seasonal and occupational personal $PM_{2.5}$ exposures were compared using analysis of variance (ANOVA). Mass differences between pairs of personal and ambient $PM_{2.5}$ data were calculated using independent sample t-test. Pearson's correlations (r) and coefficient of determination (R^2)

values were obtained to show the strength of associations between ambient and personal exposure to $PM_{2.5}$. We applied an R Squared difference test (*r2dt*) to account for the statistical differences of seasonal and occupational effects on personal-ambient associations (Jaeger, 2016). A *p-v*alue < 0.05 was considered statistically significant in a two-tailed test.

2.6 Mixed-effects models

In this analysis, a mixed-effects model was conducted in the statistical environment R 3.3.1 (R Development Core Team, 2017: http://www.r-project.org) (Bates et al., 2014). Natural logarithms were performed on all exposure data (including personal exposure to $PM_{2.5}$, OC, EC, water-soluble ions, and elements in addition to ambient $PM_{2.5}$) in mixed-effects models (1) and (2).

To take into account the variability of personal exposures, subjects were included in the model as random effects, and each exposure variable was involved as fixed effects separately in the mixed-effects model (1) (Edwards et al., 2008), which expressed as:

$$Y_{ij} = \log(X_{ij}) = \mu_Y + b_i + \varepsilon_{ij} \quad \text{Eq. 1}$$

Where μ_Y represents the fixed mean (logged) exposure level for all subjects, b_i represents the random effect associated with the i^{th} subject, and e_{ij} represents the random effect of the logged exposure level Y_{ij} associated with the i^{th} subject on the j^{th} day. In mixed-effects models, we assume that the random effects (b_i and ε_{ij}) are mutually independent with mean zero and variance components (σ^2_b and σ^2_w), respectively. Between-individual variance (σ^2_b) and within-individual variance (σ^2_w) are calculated using the method of restricted maximum likelihood (REML) (Xu, 2003).

Mixed-effects model (2) includes additional fixed effects for covariates *K* (i.e., determinants of exposure) C_1, C_2, …, C_k, which is expressed as follows:

$$Y_{ij} = \log(X_{ij}) = \mu_Y + \sum_{m=1}^{k} \beta_{mj} C_{mij} + b_i + \varepsilon_{ij} \quad \text{Eq. 2}$$

where the β_{mj} representing regression coefficients for *K* covariates. The following *K* covariates extracted from questionnaires and daily activity diaries included in mixed-effects model (2): ambient $PM_{2.5}$ concentrations at urban sites ($\mu g/m^3$), season (winter vs. summer), occupation (housewife and non-office vs. worker office worker and student), cooking (h), cleaning (h), outdoors (h), time at home (h), and time in transit (h). A mixed-effects model (2) was constructed separately for each component in $PM_{2.5}$ of personal exposures (while controlling for gender and day of the week) using a backward stepwise regression to eliminate non-significant ($p > 0.05$) variables. We use the marginal R^2 statistic (R^2_β) to measure the overall predictive ability of the mixed-effects model; a semi-partial R^2 statistic was calculated for each variable in mixed-effects model (2) (Jaeger, 2016; Jaeger et al., 2016).

3. Results

3.1 Characteristics of participants and activity profiles

Characteristics of subjects and a summary of time spent in different microenvironments associated with personal monitoring are shown in Table 1. All study subjects lived in non-smoking households, and no-ETS exposure recorded their activities during each sampling period. Few subjects indicated on their time-activity diaries any exposure to ETS during their respective sampling periods. Male (N = 25, 52.1%) and female (N = 23, 47.9%) subjects were equally represented. Keeping windows open and using air conditioner are common among study subjects (> 87.5%). Participants were categorized into four main groups including students (N = 12,

25.0%), office workers (N = 16, 33.3%), housewives (N = 12, 25.0%), and non-office workers (N = 8, 16.7%). During the summer personal monitoring period, the subjects spent 88.8% (SD = 12.2%) of their time indoors and 69.4% (SD = 22.3%) at home; during the winter, 90.9% (SD = 11.9%) indoors and 73.6% (SD = 23.1%) at home, respectively, i.e., with little difference by season (mean difference: 3-4%, $p > 0.05$). A considerable portion of time was spent at work (or in school) in summer 14.0% (standard deviation, SD = 17.8%) and winter 13.2% (SD = 17.9%). The amount of time in transit varied from 4.0% (SD = 7.8%) to 5.9% (SD = 10.0%) in winter and summer, respectively, followed by in outdoors (5.1-5.3%) and indoor cooking/dining (1.7%-3.3%). Similar results were found in previous studies in Hong Kong and other cities (Chau et al., 2002; Jahn et al., 2013; Klepeis et al., 2001; Lei et al., 2016). Graduate students in Shanghai, China spent about 86% of their time indoors, 7% in transit and 7% in outdoors (Lei et al., 2016).

3.2 Characterization of personal exposure to $PM_{2.5}$ mass and components

Table 2 reports summary statistics of ambient $PM_{2.5}$ concentrations and personal $PM_{2.5}$ mass along with their chemical components exposures. Figure 1 shows the average personal $PM_{2.5}$ exposures (ug/m^3) along with their residential locations throughout the sampling period. Average personal $PM_{2.5}$ exposures for each subject during all sampling days ranging from 9.2 $\mu g/m^3$ (SD = 0.1 $\mu g/m^3$) to 94.7 $\mu g/m^3$ (SD = 22.9 $\mu g/m^3$) (Figure 1). No significant spatial differences (p = 0.21) were found in $PM_{2.5}$ exposures for subjects living in various districts of Hong Kong in this study.

The median and mean personal $PM_{2.5}$ exposures across all subjects were 32.9 $\mu g/m^3$ and 35.4 $\mu g/m^3$ (95% confidence interval, CI: 32.4-38.4 $\mu g/m^3$), respectively. SO_4^{2-}, OC, NH_4^+, NO_3^-, and EC are the most abundant species in personal $PM_{2.5}$, all with averages that exceeded 2.2 $\mu g/m^3$. OC, EC, and water-soluble ions contributed to 24.3% (SD = 10.3%), 7.0% (SD = 2.9%), and

51.6% (SD = 14.9%) of measured personal $PM_{2.5}$ mass. The average concentrations of 19 elements (7861 ng/m^3, SD = 4775 ng/m^3) are less than the averages for carbonaceous aerosols and water-soluble ions. The mass reconstruction for personal samples was lower than personal $PM_{2.5}$ exposures obtained from the gravimetric analysis (Table 2). Strong correlations (Pearson's r: 0.96-0.97, p < 0.01) were found between the reconstruction and observation of $PM_{2.5}$ for personal exposures with a slope of 0.81 in summer and 0.85 in winter, respectively (Figure S3).

Figures 2a-b illustrate the seasonal and occupational variation of personal $PM_{2.5}$ exposures and their chemical components. Significant seasonal differences ($p < 0.01$) emerged in the average personal exposures with higher levels in winter and lower in summer for $PM_{2.5}$ mass and most ions. There was no significant seasonal fluctuation of OC and EC in personal $PM_{2.5}$ exposures. In contrast, personal exposure to Ca^{2+}, Si, Ca, and some trace elements (e.g., V, Fe, Ni, Zn) were higher in summer compared with those in winter. For most of the $PM_{2.5}$ components, significant lower exposure levels ($p < 0.05$) were found for office workers and students than other groups of subjects. In this analysis, components (Mg^{2+} and Cr) for which the percentages detected (> MDLs) lower than 60% for all samples were excluded in mixed-effects models.

3.3 Associations between personal $PM_{2.5}$ exposures and ambient concentrations

Moderate (Pearson's $r = 0.58$, $p < 0.01$) to strong (Pearson's $r = 0.65$, $p < 0.01$) personal-ambient $PM_{2.5}$ correlations were shown in Figures 3a-b. The associations varied by season ($p = 0.02$), with a slope of 0.66 (SD = 0.10) and 0.60 (SD = 0.08) in summer and winter, respectively. Figures 3c-f provide personal-ambient $PM_{2.5}$ correlations across different groups of subjects ($p < 0.01$). It is noted that stronger associations were shown for office workers (Pearson's $r = 0.69$, $p < 0.01$) and students (Pearson's $r = 0.73$ $p < 0.01$) with elevated slopes (0.60) and R^2 values.

However, moderate personal-ambient correlations with lower Pearson's r values (0.46-0.53, $p < 0.05$) and slopes (0.55-0.58) were observed for housewives and non-office workers (e.g., van drivers, paper vendors, outdoor workers).

3.4 Estimation of variance components

The between- and within-individual variance components along with variance component ratios of personal exposure to $PM_{2.5}$ mass and components on a natural log scale from mixed-effects model (1) are presented in Table 3. The within-individual variance (σ^2_w) dominated the total variability for all exposure data except $PM_{2.5}$ ($\sigma^2_b = 0.19$, 53.8%) and EC ($\sigma^2_b = 0.15$, 52.4%) (in which σ^2_b were slightly higher than σ^2_w).

Table 4 presents the variance components in mixed-effects model (2) compared with those in model (1) for all personal exposure data. Regarding the total variance components (Table 4 and Table S3), the addition of potential determinants under model (2) reduced the between-individual variance by about half for most of the $PM_{2.5}$ components in personal exposures (ranging from 48.3-87.2%), except Ca^{2+}, Ca, V, Na, Cu, and As (ranging from 25.0-38.9%). In the present study, 3 to 67 personal measurements per subject would be required for $PM_{2.5}$ mass and constituents to reduce potential attenuation bias to 20% in a hypothetical exposure-response relationship. In a previous study in Hong Kong, Pun et al. (2015) indicated that respiratory emergency hospitalizations over a consecutive six-day exposure period were the highest for vehicle exhaust (e.g., OC, EC, Ca, Fe) followed by secondary sulfate (e.g., SO_4^{2-}, NH_4^+) for PM_{10}.

3.5 Determinants of personal exposure to $PM_{2.5}$ mass and components

Table 4 summarizes the results in mixed-effects model (2), where the determinants and contributions of several variables are illustrated. Also included in Table 5 are the changes (percent change and 95% CI) for determinants in personal $PM_{2.5}$ mass and components. Table 4 shows that in model (2) the marginal R^2_β (ranging from 0.16 to 0.60) for mixed-effects model tends to be higher than the semi-partial R^2 for ambient $PM_{2.5}$ (ranging from 0.12 to 0.59) for all exposure pollutants (e.g., personal $PM_{2.5}$ mass and constituents). Among all determinants, ambient $PM_{2.5}$ dominates the contribution to personal $PM_{2.5}$ mass and components (except Cl^-, Cl, and V), with contributions from 37.3% (NO_3^-) to 99.0% (SO_4^{2-}). In this analysis, gender was considered to have no significant influence on exposure levels for all subjects. The winter season was one of the major determinants (R^2 = 0.03-0.40, $p < 0.05$) and positive contributors (5.3-70.4%) for some of the components in personal $PM_{2.5}$ (as illustrated in Table 4-5 and Table S3). As shown in model (2), occupation, time at home, outdoors, in transit, and cleaning activities were determinants of personal exposure $PM_{2.5}$ components (R^2_β = 0.06-0.63, $p < 0.05$) accounting for 3.0-29.0% of the variance. Collectively, these results indicate significant variability in personal exposure to $PM_{2.5}$ components due to individual's daily activity patterns.

4. Discussion

In the present study, personal $PM_{2.5}$ exposures among adult subjects in Hong Kong were investigated. We characterize the seasonal and occupational variations of personal exposure to $PM_{2.5}$ mass and constituents. Specifically, we examine the within- and between-individual variance components using mixed-effects models from repeated personal measurements, focusing on the determinants of individual exposures.

In this analysis, 105 out of 161 personal $PM_{2.5}$ measurements revealed concentrations above

25 μg/m^3, the recommended 24-hr ambient $PM_{2.5}$ standard issued by the World Health Organization (WHO). Average personal exposure to $PM_{2.5}$ in Hong Kong were considerably higher than those in European (ranging from 8.4 to 19.4 μg/m^3) (Johannesson et al., 2011; Lanki et al., 2007; Montagne et al., 2014) and North American cities (ranging from 12.9 to 31.4 μg/m^3) (Kim et al., 2005b; Turpin et al., 2007; Williams et al., 2000), but significantly lower than those in Chinese cities (varying from 72.6 to 126.8 μg/m^3) (Baccarelli et al., 2014; Chen et al., 2017b; Du et al., 2010; Lei et al., 2016).

Analysis of concurrent ambient and personal exposure to SO_4^{2-} and EC concentrations provides information about the estimation of individual's ambient-generated exposures (Chen et al., 2017b; Noullett et al., 2010). The results from this study showed that personal exposure to SO_4^{2-} and EC were about 1-2 orders of magnitude higher than those in the U.S. and Europe (Noullett et al., 2006; Noullett et al., 2010; Sarnat et al., 2009; Wilson and Brauer, 2006). In comparison with other studies in Chinese cities such as Guangzhou (e.g., personal exposure to sulfate and EC were 10.5 μg/m^3 (SD = 4.0 μg/m^3) and 9.7 μg/m^3 (SD = 7.3 μg/m^3) in winter) (Chen et al., 2017b), the subjects in Hong Kong were exposed to lower levels of SO_4^{2-} and EC. Analysis of elemental concentrations provides information about the corresponding sources of personal exposures (Adgate et al., 2007; Koistinen et al., 2004). Few studies have focused on measurements of personal exposure to trace elements in the general populations (Baccarelli et al., 2014; Molnár et al., 2006). Research conducted in European cities (e.g., Gothenburg, Helsinki, Utrecht, Barcelona) reported considerably lower personal elemental exposures (1.1-250 ng/m^3) compared with this study both in summer and winter (Johannesson et al., 2011; Montagne et al., 2014). An exposure study conducted in Beijing, China (Baccarelli et al., 2014), reported

372 average personal elemental exposures were 20-6190 ng/m^3 and 60-8430 ng/m^3 for office workers
373 and truck drivers, respectively.

374 Non-office workers and housewives had significantly higher ($p < 0.01$) $PM_{2.5}$ exposures
375 compared with office workers and students. Moreover, the mean subject-specific personal-to-
376 ambient $PM_{2.5}$ ratios all exceed unity (ranging from 1.1 to 1.4), highlighting the impact of non-
377 ambient generated particles on total personal exposures, especially for housewives and non-
378 office workers (Chen et al., 2017b; Noullett et al., 2010; Wilson and Brauer, 2006). Similary,
379 Williams et al. (2000) have suggested that subjects who were more sedentary may have
380 potentially lower and less variable exposures than corresponding outdoor concentrations.
381 Baccarelli et al. (2014) have reported that personal $PM_{2.5}$ exposure showed group-specific
382 profiles with significantly higher levels in truck drivers compared to office workers in Beijing,
383 China.

384 Williams et al. (2003) and Meng et al. (2009) suggested that the personal-ambient correlation
385 partially relates to differences in air exchange rate (AER). Meng et al. (2012) found that season
386 was a significant factor affecting the strength of personal-ambient $PM_{2.5}$ associations. In the
387 present study, the statistically significant differences in personal-ambient R^2 values were found
388 by season and subject. Xu et al. (2014) estimated that outdoor contributions to personal PM_{10}
389 exposures were higher in summer 55% (SD = 19%) than in winter 34% (SD = 10%) in Tianjin,
390 China. Our results agree with the findings above, on average 66% and 60% of the personal
391 exposures are due to ambient concentrations in summer and winter, respectively. The ambient
392 contribution to personal $PM_{2.5}$ exposure along with effects of seasonality on personal $PM_{2.5}$
393 exposure is further evidenced in Table 4 and 5.

394 The Pearson's correlation coefficient increased after exclusion of exposure to indoor cooking
395 (N_{sample} = 64, Pearson's r = 0.68, p < 0.01) or time in transit (N_{sample} = 87, Pearson's r = 0.74, p <
396 0.01). This is in line with previous findings, which reported improved personal-ambient
397 correlations (Spearman's r_s: 0.38 to 0.77) associated with decreased human activities (Jahn et al.,
398 2013), for example excluding ETS exposure (median, Spearman's r_s > 0.7) (Kousa et al., 2002;
399 Scapellato et al., 2009) or cooking activities (Abt et al., 2000). Although positive personal-
400 ambient relationships were shown, the lower slopes and R^2 values suggested that ambient $PM_{2.5}$
401 concentrations may not be a suitable proxy for corresponding personal exposures, especially for
402 housewife or non-office worker subjects, in cross-sectional health studies. In contrast, matched
403 pairs of daily average personal and ambient $PM_{2.5}$ concentrations yielded significant correlations
404 (Pearson's r = 0.78, p < 0.01) with a higher slope (0.73) and R^2 value (0.60). Previous findings in
405 Williams et al. (2000) and Jahn et al. (2013) have illustrated that averaging personal exposures
406 across a sub-population over time lead to improved personal-ambient $PM_{2.5}$ correlations (Jahn et
407 al., 2013; Williams et al., 2000).
408 Consideration of the relative magnitude of individual exposure variability (i.e., σ^2_w and σ^2_b)
409 can yield useful insights about optimal measurement strategy of actual exposure for study
410 subjects (Loomis and Kromhout, 2004; Weichenthal et al., 2017). For personal exposure to
411 water-soluble ions and elements, the within-individual variance (σ^2_w) accounted for a more
412 substantial part of the total variability, which is consistent with findings in previous studies
413 (Johannesson et al., 2011; Lanki et al., 2007). For instance, Lanki et al. (2007) have reported
414 relatively higher σ^2_w (ranging from 53-97%) in absorbance (i.e., as a proxy for EC) exposures
415 compared to σ^2_b (ranging from 3-63%) in indoor and personal $PM_{2.5}$. It was shown that variance
416 component ratios ($\lambda = \sigma^2_w/\sigma^2_b$) dictated the attenuation bias degree, which increases with

increasing λ, while decreases with increasing n_s (Johannesson et al., 2011). In this analysis, for personal exposure to $PM_{2.5}$ mass and most components, a reduction in σ^2_b (25.0-87.2%) and/or σ^2_w (9.2-84.2%) values were shown by adding time activity factors. According to the values of variance component ratios in model (1), the number of repeated personal samples required from each participant varied from 3 to 67, which suggests that (in the current study) personal exposure to $PM_{2.5}$ and EC would be the least biasing measures of $PM_{2.5}$ exposure for use in evaluating an exposure-response relationship. Johannesson et al. (2011) have reported that for personal exposure to $PM_{2.5}$ mass and elements, the number of repeated samples per subject to restrict attenuation bias to 20% was estimated to be 3-39. In Egeghy et al. (2005), for indoor Pb concentrations, forty-eight repeated samples per subject would be required.

Daily ambient $PM_{2.5}$, season, and occupation were significant determinants of personal exposure to $PM_{2.5}$ (R^2_β = 0.51, p < 0.0001) for all subjects throughout the study period, explaining 77.5%, 5.3%, and 13.0% of the variance, respectively. In our final model, a one-unit increase in ambient $PM_{2.5}$ (2.72 ug/m^3) was associated with a 0.75 ug/m^3 (95% CI: 0.59-0.94 ug/m^3) change in personal $PM_{2.5}$ exposure. In a previous study in Scapellato et al. (2009), researchers found that outdoor concentrations and season significantly affected personal PM_{10} exposures in asthmatic adults in Padova, Italy, contributing to 15.4% and 24.8% of the variability, respectively. In the present study, occupation was found to be a positive parameter for personal exposure to $PM_{2.5}$, EC, and Ca^{2+}, which accounted for 13.0-25.2% of the variation. Our results show an increase of 15.6% (95% CI: 3.6-28.5%), 32.5% (95% CI: 8.7-61.6%), and 41.6% (95% CI: 3.7-94.6%), respectively, for non-office workers and housewives compare with their counterparts (i.e., office workers and students) in personal exposure to $PM_{2.5}$, EC, and Ca^{2+}.

In model (2), one-unit increase in 24-hr ambient $PM_{2.5}$ associated with 0.4 ng/m^3 (95% CI:

0.8-1.5 ng/m^3) to 3.6 ng/m^3 (95% CI: 2.6-4.9 ng/m^3) change in personal exposure to the analyzed elements. Moreover, several other factors affecting personal exposure to $PM_{2.5}$ components were investigated in this study. SO_4^{2-} has been shown to be well correlated with oxalate as well as NO_3^- and NH_4^+ and is known to have limited indoor sources. Pun et al. (2014) have linked secondary nitrate (NO_3^-, NH_4^+), Na, Cl, Mg and Ni with increased hospitalization for cardiovascular and/or respiratory diseases in Hong Kong. We found that (the amount of) time spent in one's residence significantly affected personal exposure to NH_4^+, SO_4^{2-}, NO_3^-, oxalate, and S, contributing 5.6-20.9% of the variability. In this analysis, for one-hour extra time in residence (at home), an average increase of 1.7% (95% CI: 0.3%-3.2%) to 3.6% (95% CI: 1.6%-5.6%) in personal exposures were observed. Further analysis would be needed to confirm the origin (or sources) (e.g., the penetration from ambient to indoors, duration of open windows) of personal exposure to secondary ions when subjects were home. However, time at home (h/day) was found to have a negative parameter estimate for personal exposure to Ca^{2+} and Ca, contributing to 3.0-13.0% of the variability. This suggests staying at home lowered the personal Ca^{2+} exposures, indicating mostly the contribution from ambient sources rather than non-ambient ones (e.g., very local ambient sources while subjects were outdoors) (Chen et al., 2017b). It remains to be determined the associations of personal and ambient concentrations for particulate compounds.

Real-time personal monitors provide additional information on the activity pattern and peak levels of exposure (Buonanno et al., 2013; Lei et al., 2016), for example in transportation, indoor cooking, etc. Findings from our study (filter-based integrated exposures) provide direct evidence of the effect of exposure error on the ability to use ambient concentration as a proxy for personal exposure to particulate compounds, particularly those associated with individual activity patterns

463 (Chen et al., 2017a). We found time in transit associated with personal exposure to EC, NH_4^+, Ti,
464 and Fe, accounting for 19.6%, 5.6%, 7.3%, and 26.4% of the variation. Specifically, for one-hour
465 extra time in transit an average increase of 5.3% (95% CI: 1.5%-9.3%) in personal exposure to
466 EC and 2.7% (95% CI: 0.1%-10.8%) to 12.3% (95% CI: 5.6%-23.1%) in personal exposure to Ti
467 and Fe were observed, respectively. Previous studies have shown significantly higher in transit
468 EC exposures for subjects compared with time outdoors (Baccarelli et al., 2014; Kim et al.,
469 2005b; Lei et al., 2016). Time in transit was found to positively affect personal exposure to Fe (p
470 = 0.02), which was consistent with the findings in Johannesson et al. (2011). Time in transit was
471 not a significant positive estimate for $PM_{2.5}$ in mixed-effects model (2), confirming the previous
472 findings that EC is a better marker for traffic particles than $PM_{2.5}$ mass (Cyrys et al., 2003; Lei et
473 al., 2016). Baccarelli et al. (2014) reported significant higher Ti exposures (40 ng/m^3, 95% CI:
474 30-40 ng/m^3) for truck drivers during 8-h of work compared with office workers.

475 Past studies showed that cooking has often been linked with episodic peaks in $PM_{2.5}$
476 concentrations (Buonanno et al., 2013; Wallace et al., 2003). Although subjects that cooked
477 indoors were exposed to significantly higher $PM_{2.5}$ levels (8.0 μg/m^3, $p < 0.05$) than those who
478 did not cook (data not shown), cooking activity was not a significant positive contributor to 24-hr
479 $PM_{2.5}$ exposure for all subjects. The difference (8.0 μg/m^3) is in agreement with the estimate of ~
480 8 μg/m^3 in Wallace et al. (2003). In this analysis, for one-hour more spent on indoor cleaning
481 activity an average increase of 8.7% (95% CI: 0.6%-17.4%), 18.2% (95% CI: 3.3%-35.5%) and
482 19.4% (95% CI: 1.4%-40.8%) in personal exposure to NH_4^+, Mg, and V were observed,
483 respectively. Tian et al. (2013) have linked Ni and V (indicators of shipping air pollution) in
484 PM_{10} with elevated cardiovascular hospitalizations in Hong Kong. The RIOPA study indicated
485 that use of oil furnace, oven, and fireplace while indoors were possible determinants of personal

exposure to vanadium (V) (Meng et al., 2009).

One limitation of this study is the lack of concurrent indoor/outdoor (ambient) $PM_{2.5}$ constituents. Therefore no discussion concerning the homogeneity of ambient $PM_{2.5}$ components and sources contributions can be presented. Further investigations should focus on long-term monitoring better characterize total personal exposure components (ambient and non-ambient exposure) from a larger population and the corresponding health effects in epidemiological studies.

5. Conclusions

The major finding of this study confirmed that personal $PM_{2.5}$ (mass and components) exposures in Hong Kong were considerably higher than those reported in other developed countries and lower than those in Chinese cities. Significant seasonal differences ($p < 0.01$) emerged in the average personal exposures with higher levels in winter and lower levels in summer for $PM_{2.5}$ mass, most ions and elements (except Ca, Si, V, Fe, Ni, Zn). No significant seasonal variations were shown for personal exposure to OC and EC. For most personal $PM_{2.5}$ components, office workers and students had lower exposure levels than other groups of subjects. Ambient $PM_{2.5}$ concentrations may not be a reasonable proxy for personal exposures in housewives or non-office workers, and further investigation into relationships of ambient concentrations with the corresponding total exposure components (i.e., ambient and non-ambient origin) is warranted to elucidate the health risks associated with $PM_{2.5}$ exposure in epidemiological studies. Aside from ambient concentration, seasonality and occupation, individual activities (time at home, outdoors, time spent in transit, and cleaning activities) were significant determinants of personal exposure to OC, EC, major ions, and trace elements (including Ti, V, and Fe). We found that the within-

individual variance component dominated the total variability for most of the particulate species, which point to the importance of obtaining repeated samples from study subjects in improving epidemiological associations. Our study highlights the need for conducting personal monitoring along with time activity survey to elucidate determinants of individual's exposures and develop effective exposure mitigation strategies.

Conflicts of interest

The authors declare no conflict of interest.

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Tables

Table 1. Description detail and subjects' activity during the personal sampling campaign.

	Summer	Winter	Total
Sampling Date	July – October 2014	December 2014- March 2015	
Study subjects (N)	42 (35[a])	41 (35 [a])	48
Gender			
Female	20	19	23 (47.9%)
Male	22	22	25 (52.1%)
Age, median (range)	28 (18-63)	27 (18-63)	
18-20			7 (15.6%)
20-40			25 (55.6%)
40-65			13 (28.9%)
Occupation (N, %)			
Student	14	16	16 (33.3%)
Office worker	10	10	12 (25.0%)
Housewife	10	10	12 (25.0%)
Non-office worker[b]	8	5	8 (16.7%)
Smokers (Yes/No, N, %)			No (48, 100%)
ETS[c] at home, indoors (Yes/No, N, %)			No (48, 100%)
Air condition use (Yes/No, N[d], %)			Yes (42, > 87.5%)
Open windows (Yes/No, N[d], %)			Yes (42, > 87.5%)
Energy for cooking (N[d], %)			
Gas stove			12 (28.6%)
Town gas			13 (31.0%)
LPG[e]			5 (11.9%)
Electricity			5 (11.9%)
No cooking energy available			7 (16.7%)
Time-activity data from diaries (%)			
Time spent indoors	Median (Mean, SD[f])	Median (Mean, SD[f])	
Indoors, total	92.2% (88.8%, 12.2%)	93.8% (90.9%, 11.9%)	
Indoors, at home	71.9% (69.4%, 22.3%)	79.2% (73.6%, 23.1%)	
Indoors, work/school	0% (14.0%, 17.8%)	0% (13.2%, 17.9%)	
Kitchen (Cooking/Dining)	4.2% (4.7%, 4.6%)	6.3% (7.5%, 7.3%)	
Cleaning activities[e]	0% (1.7%, 3.0%)	0% (3.3%, 5.5%)	
Outdoors	3.1% (5.3%, 7.0%)	2.8% (5.1%, 9.5%)	
Transportation (Metro, bus/minibus)	3.6% (5.9%, 10.0%)	0% (4.0%, 7.8%)	

Notes: [a]Number of recruited subjects participated both in summer and winter sampling campaign. [b]Technicians, divers, paper vendors, van drivers. [c]Exposure to environmental tobacco smoke (ETS). [d]Data not avaiable for six subjects. [e]LPG denotes liquefied petroleum gas. [e]Dusting, cleaning, and vacuuming. [f]SD denotes standard deviation.

Table 2. Descriptive statistics for 24 h ambient $PM_{2.5}$ mass concentrations at urban sites, in addition to personal exposure to $PM_{2.5}$, carbonaceous materials, water-soluble ions, and trace elements in the general population of Hong Kong.

		Mean	SD[a]	Median	Min-Max[b]	IQR[c]	95% CI[d]	N[e]	MDL[f]	> MDL[f] (%)
$PM_{2.5}$ (μg/m^3)	Personal exposure	35.4	19.5	32.9	3.5-110.9	25.2	32.4-38.4	161	0.33	100
	Ambient level[#]	35.3	19.4	37.6	5.8-105.3	25.2	32.3-38.3	161	N.A.	100
	Reconstructed mass[g]	33.0	18.4	30.0	5.5-93.9	21.5	30.1-35.8	161	N.A.	100
Carbonaceous materials (μg/m^3)	OC	7.8	4.9	6.6	2.5-40.8	4.7	7.1-8.6	161	0.28	100
	EC	2.2	1.1	2.1	0.5-5.2	1.6	2.0-2.4	161	0.04	100
Water-soluble ions (μg/m^3)	Na^+	0.6	0.3	0.5	0.2-1.9	0.3	0.5-0.6	155	0.18	96.3
	NH_4^+	4.2	2.7	3.8	0.3-12.8	3.3	3.7-4.6	157	0.23	97.5
	K^+	0.3	0.2	0.3	0.1-1.3	0.3	0.3-0.4	151	0.01	93.8
	Mg^{2+}	0.1	0.0	0.1	0.1-0.2	0.1	0.10-0.11	82	0.02	50.9
	Ca^{2+}	0.3	0.6	0.2	0.1-6.0	0.2	0.2-0.4	151	0.03	93.8
	Cl^-	0.4	0.5	0.2	0.1-3.2	0.4	0.3-0.5	153	0.03	95.0
	NO_3^-	3.0	3.9	1.6	0.1-23.4	2.7	2.4-3.6	161	0.01	100
	SO_4^{2+}	9.8	5.8	9.3	0.9-26.6	7.3	8.9-10.7	161	0.01	100
	Oxalate	0.4	0.8	0.3	0.0-7.2	0.3	0.3-0.5	139	0.01	86.3
Elements (ng/m^3)	Na	3482	2171	3321	50-9095	2776	3140-3824	155	33	96.3
	Mg	150	101	125	18-422	142	131-169	107	1	66.5
	Al	139	114	126	9-771	150	120-157	148	5	91.9
	Si	259	384	171	4-3759	244	199-319	156	3	96.9
	S	2757	1549	2751	84-7503	2197	2518-2996	161	2	100
	Cl	187	308	61	6-2140	158	139-235	156	5	96.9
	K	331	248	296	8-1288	293	293-369	161	3	100
	Ca	290	1093	142	9-12167	153	120-459	160	2	99.4
	Ti	15	17	12	2-159	11	12-17	156	1	96.9
	V	15	16	8	1-77	13	12-17	156	1	96.9
	Cr	3	2	3	0.9-11	3	3-4	86	0.9	53.4
	Mn	14	13	13	0.8-133	11	12-16	151	0.8	93.8
	Fe	302	326	213	1-2655	269	252-353	161	0.7	100
	Ni	5	4	3	0.5-22	4	4-5	143	0.5	88.8
	Cu	23	23	18	0.9-138	18	20-27	154	0.5	95.7
	Zn	134	219	104	2-2456	126	100-168	161	0.5	100
	As	3	2	3	0.8-9	2	3-4	103	0.8	64.0
	Br	15	12	11	0.7-67	13	13-16	157	0.5	97.5
	Pb	29	22	26	2-97	32	26-33	138	0.5	85.7

Notes: [a]SD refers to standard deviation. [b]Min, Minimum; Max, Maximum. [c]IQR refers to the interquartile range. [d] 95% confidence interval for the mean of the individual's exposure and the level of significance was taken as $p < 0.05$. [e]N refers to the number of valid analytical results. [f]MDL refers to method detection limit; concentrations below the detection limit were discarded. [f][Reconstructed Mass] for personal exposure = (1.89 ×[Al] + 2.14×[Si] +1.4 ×[Ca] +1.43 ×[Fe]) + (1.4 ×[OC] + [EC]) + (1.38 ×[SO42-] + 1.29 ×[NO3-]) + [Na+] + non-crustal elements excluding geological material (e.g, Al, Si, Ca, Fe, S).

[#]Spearman's r_s ranged from 0.78 to 0.95 between eleven urban air quality monitoring stations (including Central/Western, Eastern, Kwai Chung, Kwun Tong, Sham Shui Po, Tsuen Wan, Sha Tin, Tai Po, Tuen Mun, Tung Chung, Yuen Long). 24-hr average ambient $PM_{2.5}$ (summer: 21.3 μg/m^3 (SD = 15.6 μg/m^3) ~ 34.7 μg/m^3 (SD = 14.3 μg/m^3); winter: 36.5 μg/m^3 (SD = 16.0 μg/m^3) ~ 51.8 μg/m^3 (SD = 26.9 μg/m^3) from all these sites were compared with personal $PM_{2.5}$. N.A. denotes not available.

Table 3. Parameters estimated from mixed-effects model (1) for personal exposure to $PM_{2.5}$, carbonaceous materials, water-soluble ions, and the trace elements from the study subjects.

		Subjects (n_s)	Samples (N)	σ^2_b	σ^2_w	σ^2_b (%)	σ^2_w (%)	> MDL[a] (%)	λ[b]	n[c]
	Personal exposure to $PM_{2.5}$	48	161	0.19	0.16	53.8	46.2	100	0.9	3
Carbonaceous materials	OC	48	161	0.12	0.15	44.1	55.9	100	1.3	5
	EC	48	161	0.15	0.14	52.4	47.6	100	0.9	4
Water-soluble ions	Na^+	48	155	0.00	0.21	- [d]	100.0	96.3	- [d]	- [d]
	NH_4^+	48	157	0.24	0.34	41.5	58.5	97.5	1.4	6
	K^+	47	151	0.14	0.28	34.4	65.6	93.8	1.9	8
	Ca^{2+}	48	151	0.26	0.28	48.4	51.6	93.8	1.1	4
	Cl^-	48	153	0.00	0.78	- [d]	100.0	95.0	- [d]	- [d]
	NO_3^-	48	161	0.48	0.63	43.3	56.7	100	1.3	5
	SO_4^{2+}	48	161	0.22	0.31	42.0	58.0	100	1.4	6
	oxalate	46	139	0.16	0.42	27.0	73.0	86.3	2.7	11
Elements	Na	48	155	0.09	0.36	43.9	56.1	96.3	1.3	5
	Mg	47	107	0.04	0.28	13.9	86.1	65.2	6.2	25
	Al	48	148	0.04	0.75	5.6	94.4	75.8	16.9	67
	Si	48	156	0.31	0.92	25.5	74.5	93.2	2.9	12
	S	48	161	0.15	0.28	34.7	65.3	97.5	1.9	8
	Cl	48	156	0.12	0.27	30.9	69.1	96.9	2.2	9
	K	48	161	0.06	0.19	23.8	76.2	97.5	3.2	13
	Ca	48	160	0.24	0.59	28.8	71.2	99.4	2.5	10
	Ti	48	156	0.72	0.93	43.7	56.3	96.9	1.3	5
	V	48	156	0.37	0.68	35.3	64.7	95.7	1.8	7
	Mn	48	151	0.09	0.80	10.0	90.0	65.8	9.0	36
	Fe	48	161	0.15	0.27	35.5	64.5	93.8	2.3	9
	Ni	47	143	0.19	0.37	34.3	65.7	89.4	1.9	8
	Cu	48	154	0.05	0.20	19.6	80.4	88.2	4.1	16
	Zn	48	161	0.32	0.96	24.9	75.1	95.0	3.0	12
	As	43	103	0.04	0.35	10.2	89.8	64.6	8.8	35
	Br	48	157	0.33	0.66	33.5	66.5	78.9	2.0	8
	Pb	48	138	0.01	0.16	3.1	96.9	82.6	5.3	21

Notes: σ^2_b, between-individual variance. σ^2_w, within-individual variance. ICC = $\sigma^2_b / (\sigma^2_b + \sigma^2_W)$, denotes proportion of the variation attributed to between-individual variance. [a]MDL refers to method detection limit, [b]$\lambda = \sigma^2_W / \sigma^2_b$, [c]Number of repeated samples from each subject to reduce attenuation bias to 20%, (e.g., n = 4*λ, which has been described in detail by Johannesson, Rappaport et al. (2011)). [d]Could not estimated.

35

36 **Table 4.** Parameters estimated from mixed-effects model (2) for $PM_{2.5}$, OC, EC, ions, and the trace elements based on personal samples from the study subjects.

	Subjects	Samples	Fixed effects	Estimate	*p*-value	R^2_β	Contribution	Model 2			Reduction	
	n_s	N					(%)[b]	σ^2_b	σ^2_w	ICC (%)	σ^2_b (%)	σ^2_w (%)
$PM_{2.5}$	48	156	Intercept	1.35	< 0.0001	**0.51**[a]		0.08	0.09	47.1	57.5	44.3
			Ambient concentration	0.56	< 0.0001	0.39	77.5					
			Season	0.15	0.009	0.03	5.3					
			Occupation	0.11	0.02	0.07	13.0					
OC	48	156	Intercept	0.48	0.01	**0.28**[a]		0.06	0.12	33.3	48.5	18.9
			Ambient concentration	0.41	< 0.0001	0.26	94.6					
			Outdoor	0.05	0.01	0.08	29.0					
EC	48	156	Intercept	-0.81	< 0.0001	**0.40**[a]		0.08	0.10	44.4	48.3	28.9
			Ambient concentration	0.39	< 0.0001	0.24	59.4					
			Occupation	0.28	0.007	0.09	21.5					
			In transit	0.05	< 0.0001	0.05	19.6					
Na^+	48	150	Intercept	-1.17	< 0.0001	**0.04**[a]		0.001	0.20	0.5	-[c]	3.3
			Ambient concentration	0.12	0.02							
NH_4^+	47	153	Intercept	-2.19	< 0.0001	**0.63**[a]		0.05	0.17	22.7	79.3	50.1
			Ambient concentration	0.80	0.0002	0.52	82.8					
			Season	0.27	0.0002	0.08	12.7					
			In transit	0.04	0.05	0.05	8.6					
			Indoors, at home	0.02	0.004	0.04	5.6					
			Cleaning	0.08	0.04	0.03	5.2					
K^+	47	146	Intercept	-3.89	< 0.0001	**0.45**[a]		0.05	0.19	20.8	65.5	31.2
			Ambient concentration	0.742	< 0.0001							
Ca^{2+}	48	146	Intercept	-2.85	< 0.0001	**0.25**[a]		0.19	0.16	52.4	27.9	28.9
			Ambient concentration	0.44	< 0.0001	0.13	54.5					
			Occupation	0.35	0.03	0.06	25.2					
			Indoors, at home	-0.02	0.01	0.03	13.0					
Cl^-	48	148	Intercept	-1.79	< 0.0001	**0.20**[a]		0.01	0.71	0.7	-[c]	9.1
			Season	0.85	< 0.0001							
NO_3^-	48	156	Intercept	-2.84	< 0.0001	**0.56**[a]		0.15	0.42	54.3	68.7	43.1
			Ambient concentration	0.45	< 0.0001	0.21	37.3					
			Season	1.28	< 0.0001	0.40	70.4					
			Indoors, at home	0.03	0.03	0.03	5.7					
SO_4^{2-}	48	156	Intercept	-1.12	< 0.0001	**0.60**[a]		0.03	0.16	15.8	86.5	47.8
			Ambient concentration	0.85	< 0.0001	0.59	99.0					
			Indoors, at home	0.02	0.01	0.04	6.7					
oxalate	46	136	Intercept	-4.67	< 0.0001	**0.44**[a]		0.02	0.31	6.1	87.2	26.6
			Ambient concentration	0.80	< 0.0001	0.40	91.3					
			Indoors, at home	0.04	0.0004	0.09	20.9					
Mg	47	107	Intercept	-8.17	< 0.0001	**0.06**[a]		0.02	0.09	18.2	55.5	67.6
			Cleaning	0.17	< 0.0001							
S	48	161	Intercept	-12.32	< 0.001	**0.59**[a]		0.04	0.09	30.8	73.0	67.6

			Ambient concentration	0.96	< 0.0001	0.59	99.0						
			Indoors, at home	2.79	< 0.0001	0.03	5.7						
K	48	161	Intercept	-10.35	< 0.0001	**0.59**[a]		0.01	0.03	25.0	83.1	84.2	
			Ambient concentration	0.51	0.010	0.51	86.8						
			Season	0.08	< 0.0001	0.08	14.2						
Ca	48	161	Intercept	-11.30	< 0.0001	**0.16**[a]		0.15	0.43	25.9	37.6	27.6	
			Ambient concentration	0.82	< 0.0001	0.12	75.0						
			Indoors, at home	-0.59	< 0.0001	0.005	3.0						
Ti	48	156	Intercept	-10.32	< 0.0001	**0.37**[a]		0.32	0.39	45.1	55.5	58.0	
			Ambient concentration	0.35	< 0.0001	0.35	94.3						
			In transit	0.03	0.05	0.03	7.3						
V	48	156	Intercept	-12.72	< 0.0001	**0.03**[a]		0.28	0.62	31.1	25.0	9.2	
			Cleaning	0.18	< 0.0001								
Fe	48	161	Intercept	13.96	< 0.0001	**0.33**[a]		0.03	0.11	21.4	80.0	59.5	
			Ambient concentration	0.89	< 0.0001	0.27	80.6						
			In transit	0.13	0.007	0.09	26.4						

Notes: [a]The marginal R^2 statistic for the overall mixed-effects model are marked in bold (R^2_β). [b]Denotes percentage of variance (Contribution = R^2/ R^2_β *100%) calculated for each fixed effect in the model. [C]Could not be estimated. σ^2, estimated variance of log-transformed concentrations; σ^2_b, between-individual variance, and σ^2_w, within-individual variance. Contribution (%) = Reduction (%) = $([\sigma^2_{T1} - \sigma^2_{T2}] *100/ \sigma^2_{T1})$, where $\sigma^2_T = \sigma^2_w + \sigma^2_b$.

Table 5. Effects (change and 95% confidence interval (CI)) of determinants on personal exposure to $PM_{2.5}$ mass, OC[#], EC, ions, and the trace elements.

	Change[a] (95% CI)		% Change[b] (95% CI)				R^2_β
	Ambient $PM_{2.5}$ (e)[d] +	Season (winter vs. summer)	Occupation[e]	Indoors, at home (1h/day) +	In transit (1 h/day) +	Cleaning (1 h/day) +	
$PM_{2.5}$ (ug/m^3)	0.75 (0.59-0.94)	0.16 (0.04-0.29)	15.6% (3.6-28.5%)	-[c]	-[c]	-[c]	0.51*
OC (ug/m^3)	0.51 (0.36-0.67)	-[c]	-[c]	-	-	-	0.28*
EC (ug/m^3)	0.47 (0.34-0.63)	-	32.5% (8.7-61.6%)	-	5.3% (1.5-9.3%)	-	0.40**
NH_4^+ (ug/m^3)	1.23 (0.96-1.54)	0.31 (0.14-0.51)	-	2.4% (0.8-4.0%)	4.4% (0.1-8.9%)	8.7% (0.6-17.4%)	0.63*
Ca^{2+} (ug/m^3)	0.55 (0.33-0.81)	-	41.6% (3.7-94.6%)	-2.2% (-3.9%, 0.5%)	-	-	0.25*
NO_3^- (ug/m^3)	0.57 (0.55-1.37)	2.60 (1.86-3.53)	-	2.6% (0.3-5.1%)	-	-	0.56*
SO_4^{2-} (ug/m^3)	1.34 (1.08-1.64)	-	-	1.7% (0.3-3.2%)	-	-	0.60*
oxalate (ug/m^3)	1.23 (0.94-1.64)	-	-	3.6% (1.6-5.6%)	-	-	0.44**
Mg (ng/m^3)	-[c]	-	-	-	-	18.2% (3.3-35.5%)	0.06**
Si (ng/m^3)	2.0 (1.4-2.8)	0.9 (0.5-1.5)	-	-	-	-	0.43*
S (ng/m^3)	1.6 (1.3-2.0)	-	-	1.8% (0.2-3.5%)	-	-	0.59**
K (ng/m^3)	1.7 (1.3-3.1)	0.4 (0.2-0.8)	-	-	-	-	0.59**
Ca (ng/m^3)	0.7 (0.4-1.0)	-	-	-3.6% (-5.8%, -1.5%)	-	-	0.16**
Ti (ng/m^3)	0.4 (0.8-1.5)	-	-	-	2.7% (0.1-10.8%)	-	0.37*
V (ng/m^3)	-	-	-	-	-	19.4% (1.4-40.8%)	0.03**
Mn (ng/m^3)	1.4 (1.0-1.9)	-	-	-	-	-	0.38**
Fe (ng/m^3)	1.4 (0.9-2.1)	-	-	-	12.3% (5.6-23.1%)	-	0.33**
Cu (ng/m^3)	0.9 (0.5-1.4)	-	-	-	-	-	0.17*
Zn (ng/m^3)	2.4 (1.8-3.2)	-	-	-	-	-	0.45*
As (ng/m^3)	1.4 (0.9-2.1)	-	-	-	-	-	0.33**
Br (ng/m^3)	1.7 (1.3-2.2)	0.6 (0.3-0.9)	-	-	-	-	0.60**
Pb (ng/m^3)	3.6 (2.6-4.9)	-	-	-	-	-	0.50**

*$\alpha = 0.05$; **$\alpha = 0.01$. The estimated effects for determinants are presented as: a) change $[exp^{estimate} - 1]$ and b) percentage change $[(exp^{estimate} - 1) \times 100\%]$. [c]Variable not considered a potential covariate for exposure pollutants. [d]e ≈ 2.72 ug/m^3. [e]Housewife and non-office worker vs. office worker and student. [#]For one-hour more spent outdoors an average increase of 4.7% (95% CI, 1.0-8.6%) in personal exposure to OC was observed.

Figure Captions

Figure 1. Subjects' residential locations along with average personal $PM_{2.5}$ exposures (ug/m^3) in the general population of Hong Kong during July 2014-March 2015.

Figure 2. Characterization of personal exposure to $PM_{2.5}$ along with the components in $PM_{2.5}$ of personal exposures in Hong Kong by (a) season and (b) groups of subjects.

Figure 3. Relationships between personal $PM_{2.5}$ exposures and corresponding ambient $PM_{2.5}$ concentrations at urban sites during (a) summer and (b) winter; Relationships between personal $PM_{2.5}$ exposures and ambient $PM_{2.5}$ across (c) office workers, (d) students, (e) housewives, and (e) non-office workers throughout the study period. *The difference is significant at the 0.05 level; **The difference is significant at the 0.01 level.

Figures

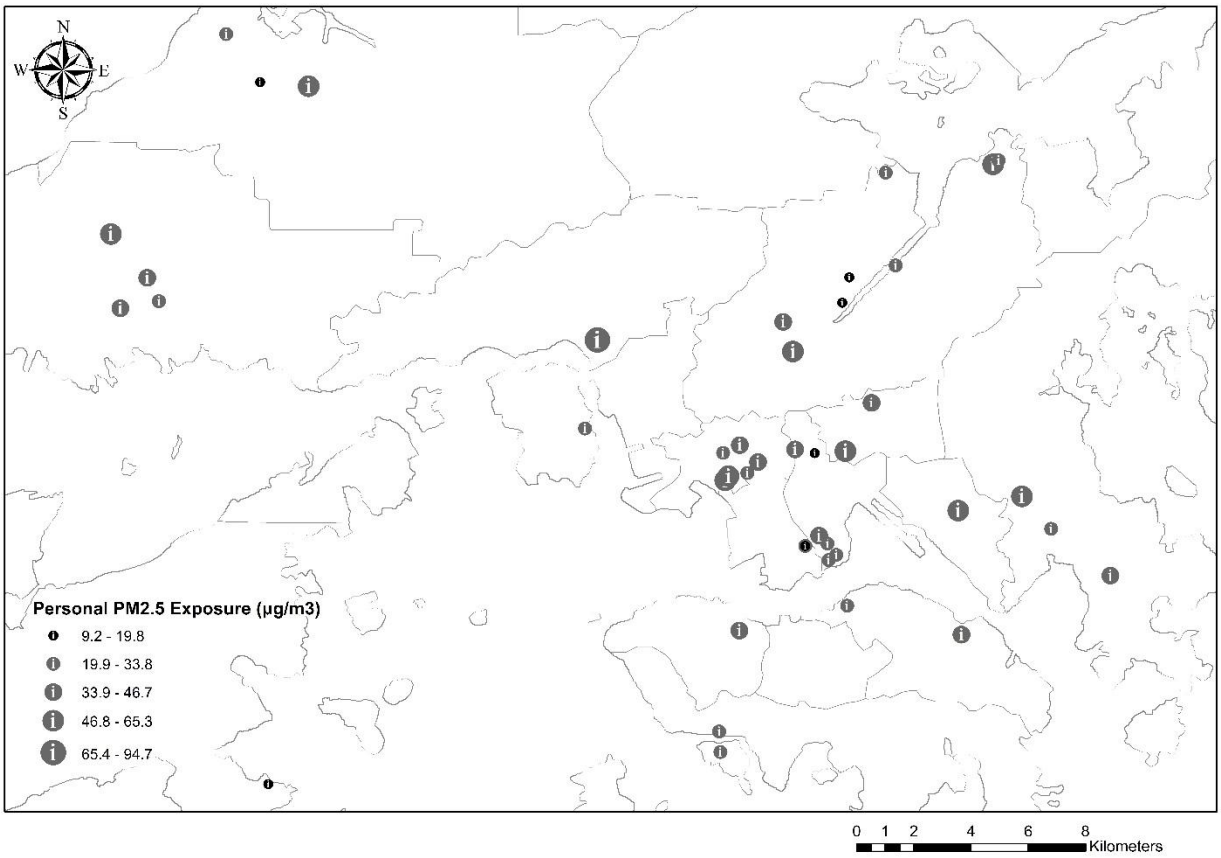

Figure 1

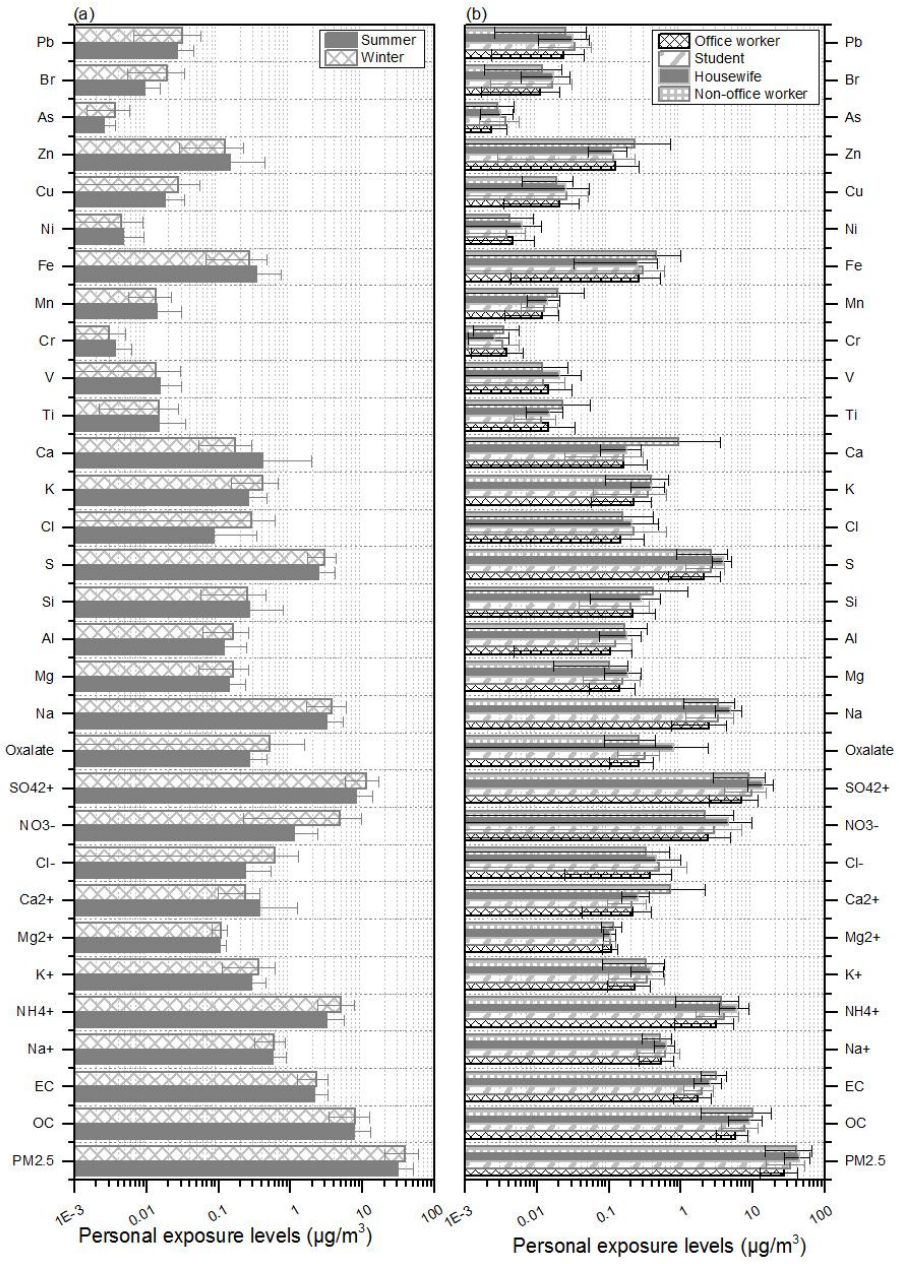
(a)
(b)
Summer
Winter
Office worker
Student
Housewife
Non-office worker
Pb
Br
As
Zn
Cu
Ni
Fe
Mn
Cr
V
Ti
Ca
K
Cl
S
Si
Al
Mg
Na
Oxalate
SO42+
NO3-
Cl-
Ca2+
Mg2+
K+
NH4+
Na+
EC
OC
PM2.5
1E-3
0.01
0.1
1
10
100
Personal exposure levels (μg/m3)

Figure 2

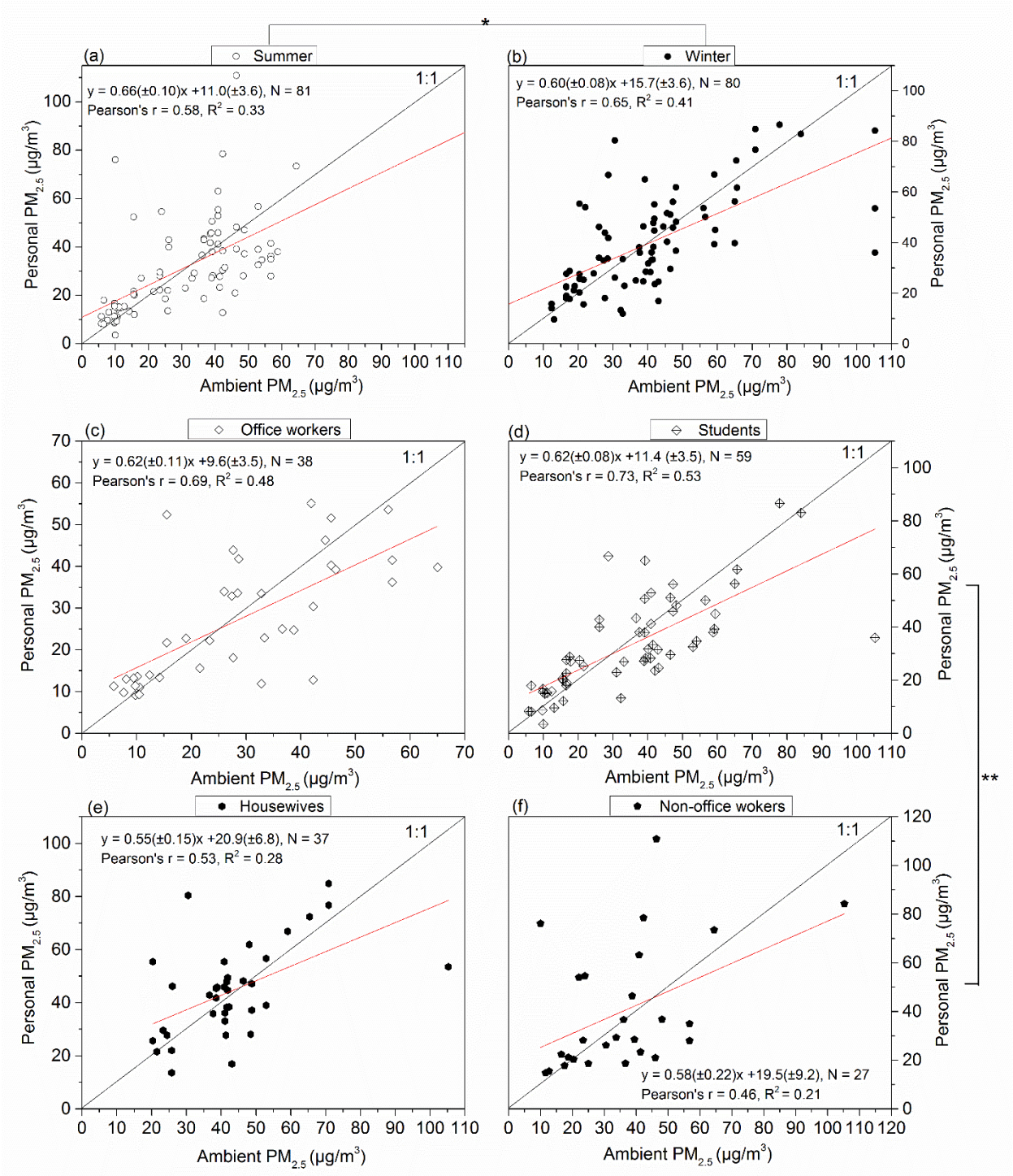
*
(a)
Summer
y = 0.66(±0.10)x +11.0(±3.6), N = 81
Pearson's r = 0.58, R2 = 0.33
1:1
(b)
Winter
y = 0.60(±0.08)x +15.7(±3.6), N = 80
Pearson's r = 0.65, R2 = 0.41
1:1
(c)
Office workers
y = 0.62(±0.11)x +9.6(±3.5), N = 38
Pearson's r = 0.69, R2 = 0.48
1:1
(d)
Students
y = 0.62(±0.08)x +11.4 (±3.5), N = 59
Pearson's r = 0.73, R2 = 0.53
1:1
(e)
Housewives
y = 0.55(±0.15)x +20.9(±6.8), N = 37
Pearson's r = 0.53, R2 = 0.28
1:1
(f)
Non-office wokers
y = 0.58(±0.22)x +19.5(±9.2), N = 27
Pearson's r = 0.46, R2 = 0.21
1:1
Personal PM2.5 (μg/m3)
Ambient PM2.5 (μg/m3)
**

Figure 3

Supplementary material for on-line publication only

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30th January 2018

Author agreement

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