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Estimation of personal exposure to fine particles ($PM_{2.5}$) of ambient origin for healthy adults in Hong Kong

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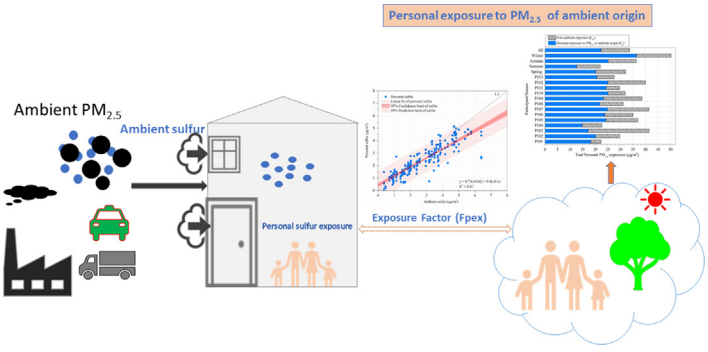
HIGHLIGHTS

- A wide range of trace elements were investigated for a year-long ambient and personal $PM_{2.5}$ samples.
- An exposure factor of 0.73 ± 0.02 was estimated using ambient $PM_{2.5}$ sulfur as a surrogate.
- $PM_{2.5}$ of ambient origin (E_a) accounted for ~57–73% of total personal exposure by season.
- Using ambient $PM_{2.5}$ at central monitoring stations as proxies underestimates the true exposures by 16–28%.

GRAPHICAL ABSTRACT



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ABSTRACT

Personal exposure and ambient fine particles ($PM_{2.5}$) measurements for 13 adult subjects (ages 19–57) were conducted in Hong Kong between April 2014 and June 2015. Six to 21 personal samples (mean = 19) per subject were obtained throughout the study period. Samples were analyzed for mass by gravimetric analysis, and 19 elements (from Na to Pb) were analyzed using X-Ray Fluorescence. Higher subject-specific correlations between personal and ambient sulfur ($r_s = 0.92$; $p < 0.001$) were found as compared to $PM_{2.5}$ mass ($r_s = 0.79$; $p < 0.001$) and other elements ($0.06 < r_s < 0.86$). Personal vs. ambient sulfur regression yielded an average exposure factor (F_{pex}) of 0.73 ± 0.02, supporting the use of sulfur as a surrogate to estimate personal exposure to $PM_{2.5}$ of ambient origin (E_a). E_a accounted for 41–82% and 57–73% of total personal $PM_{2.5}$ exposures (P) by season and by subject, respectively. The importance of both E_a and non-ambient exposures (E_{na}, 11.2 ± 5.6 μg/m^3; 32.5 ± 10.9%) are noted. Mixed-effects models were applied to estimate the relationships between ambient $PM_{2.5}$ concentrations and their corresponding exposure variables (E_a, P). Higher correlations for E_a (0.90; $p < 0.001$) than for P (0.58; $p < 0.01$) were found. A calibration coefficient < 1 suggests an attenuation of 22% (ranging 16–28%) of the true effect estimates when using average ambient concentrations at central monitoring stations as surrogates for E_a. Stationary ambient data can be used to assess population exposure only if PM exposure is dominated by E_a.

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1. Introduction

Epidemiological studies show that elevated $PM_{2.5}$ and PM_{10} (particles with aerodynamic diameters <2.5 and 10 μm, respectively) concentrations are associated with cardiovascular and respiratory morbidity and mortality (Boldo et al., 2006; Brook et al., 2010; Franklin et al., 2006; Nel, 2005). Associations of short-term and long-term exposure to $PM_{2.5}$ and PM_{10} mass and their components with daily cardiovascular and respiratory emergency hospitalizations and mortality were reported in Hong Kong (Pun et al., 2014; Wong et al., 2015), confirming the adverse health effects of $PM_{2.5}$ (Ruckerl et al., 2011).

Since most people spent ~80–90% of their time indoors (Chen et al., 2018; Jahn et al., 2013; Klepeis et al., 2001), personal $PM_{2.5}$ exposures (P) can differ from those of ambient concentrations (C) measured at central monitoring stations (Sarnat et al., 2010). Cross-sectional studies showed weak personal-ambient $PM_{2.5}$ correlations (Lachenmyer, 2000; Meng et al., 2004; Oglesby et al., 2000), with stronger associations reported in longitudinal studies (Adgate et al., 2003; Rhomberg et al., 2011; Sarnat et al., 2000). Zeger et al. (2000) provided a statistical system to estimate the effects of measurement error on health risk estimates. Several types of exposure measurement error (e.g., classical error, Berkson error) have been investigated (Dionisio et al., 2016; Goldman et al., 2010; Rhomberg et al., 2011). These studies indicated that Berkson error increases the variance of regression coefficients, while the classical error was influenced by indoor sources and particles generated from personal activities (Koenig et al., 2005; Zeger et al., 2000). These errors may cause bias in air pollution epidemiology (Avery et al., 2010; Goldman et al., 2011; Kioumourtzoglou et al., 2014).

Some studies have used sulfate and/or sulfur as tracers to estimate personal exposure to $PM_{2.5}$ of ambient origin (E_a) based on the assumptions that sulfate/sulfur compounds are primarily originated from outdoor pollution sources (Chen et al., 2017; Sarnat et al., 2009; Sarnat et al., 2002; Wallace and Williams, 2005). Other PM elements (e.g., nickel, iron) have also been utilized as surrogates for ambient $PM_{2.5}$ (Ji et al., 2018; Long and Sarnat, 2004). Further, the exposure factor (F_{pex}) has been used to estimate the outdoor (ambient) contributions to total personal exposure (Meng et al., 2005; Rhomberg et al., 2011); F_{pex} varies with seasons, individuals, and geographic regions (Sarnat et al., 2009; Wallace and Williams, 2005). These studies have reported higher P-C sulfate/sulfur correlations than for $PM_{2.5}$ mass (Ebelt et al., 2005; Noullett et al., 2010).

Associations between health outcomes and exposure components (i.e., E_a and non-ambient exposures [E_{na}]) also characterize the exposure-epidemiological relationships (Ebelt et al., 2005; Ji and Zhao, 2015; Meng et al., 2005; Wilson and Brauer, 2006). Some of these studies concluded that the strength of association between ambient concentrations and true exposures tended to bias the health effect estimates towards the null (Dominici et al., 2000; Schwartz et al., 2007; Strand et al., 2005). Therefore, regression coefficients between ambient $PM_{2.5}$ concentrations and true exposures (e.g., P, E_a) were used to evaluate the bias in $PM_{2.5}$-mediated health effects (Avery et al., 2010; Kioumourtzoglou et al., 2014).

Past studies in Hong Kong have investigated ambient and/or indoor PM mass and chemical concentrations (Ho et al., 2004; Huang et al., 2013; Wong et al., 2016). Other studies have focused on the health risks of source-specific $PM_{2.5}$, with a limited number of studies conducted that have evaluated the characteristics of personal exposures or the related health risks (Fan et al., 2018). The work reported here investigates the relationships between ambient concentrations and corresponding personal $PM_{2.5}$ mass and elements as well as addressing the utility of sulfur as an estimator for E_a. The magnitudes of exposure measurement error are examined, and correlations between the estimated exposures (e.g., E_a) and measured parameters (i.e., P and C) are assessed using mixed-effects models.

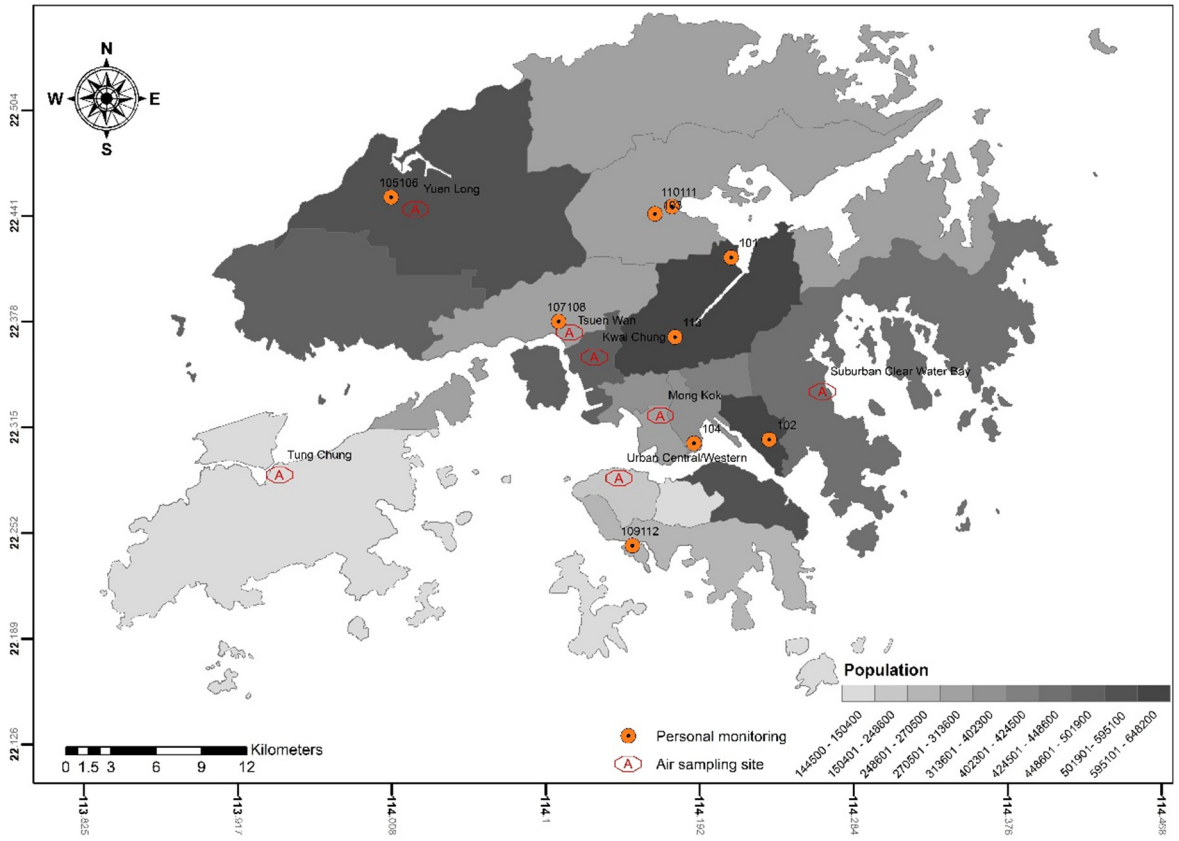

Fig. 1. Map of the study area in Hong Kong with seven air quality monitoring stations (labeled A) and study subjects' residences for personal monitoring (orange circles).

2. Material and methods

2.1. Study design and sampling methods

Thirteen adult subjects (ages 19–57) living and working in different Hong Kong districts participated in this investigation (Fig. 1). All subjects were non-smokers, were not exposed to environmental tobacco smoke in indoor microenvironments (e.g., home, school, office, or other indoors), and did not have any chronic diseases. Sampling was conducted between April 2014 and June 2015. Eleven of the thirteen subjects completed the four sampling seasons; one subject participated from April to August 2014 and another one from October 2014 to June 2015. The Joint Chinese University of Hong Kong-New Territories East Cluster Clinical Research Ethics Committee (Ref. No. CRE-2014.154) approved this study before subject recruitment. A written informed consent was obtained from each subject before that subject participated in this study.

2.1.1. Personal monitoring

Twenty-four hour (24-h, 00:00–24:00 local time) personal $PM_{2.5}$ exposure was measured using a Personal Environmental Monitor (PEM, Model 200, MSP Corp., Shoreview, MN, USA) with a Leland Legacy pump (SKC Inc., Eighty-Four, PA, USA) operated at a flow rate of 10 L/min. The PEM was loaded with a 37 mm Teflon membrane filter (PTFE, 2 μm pore size, PALL) and worn near each subject's breathing zone during sampling. Subjects were required to carry the sampler at all times except for sleeping, sitting, or other activities when the sampler was placed in close proximity (<1 m from the subject's breathing zone) and at the same height as them.

The 13 subjects were divided into three groups, sampling once 18th day for each group. This resulted in 6 to 21 personal samples per participant, which provided a longitudinal record. Samples were excluded from the analysis if the total sampling time was <16-h (due to pump failure) and/or if the filter was contaminated. Out of the 248 personal samples, 242 exposure events (97.6%) passed the quality checks for follow-up data analyses.

Each participant was required to complete a general questionnaire regarding personal information before participating in the study. Study subjects were encouraged to maintain regular daily activity patterns. During each monitoring session, subjects were also required to fill out a 24-h time-activity diary. Additional details about personal monitoring can be found in our recent publication (Chen et al., 2018).

2.1.2. Concurrent ambient monitoring

Twenty-four hour (00:00 to 24:00 local time) ambient $PM_{2.5}$ samples were collected every-sixth day at seven central monitoring stations using Partisol Samplers (Model 2025i, Thermal Fisher Scientific Inc., MA, USA) equipped with $PM_{2.5}$ inlets operated at a flow rate of 16.7 L/min (http://www.aqhi.gov.hk/en.html). A total of 467 ambient $PM_{2.5}$ samples were acquired over the year-long sampling period. Daily meteorological data, including ambient pressure (P), temperature (T), relative humidity (RH), wind speed (WS), wind direction (WD), and rainfall (R) from the Hong Kong Observatory (HKO, http://www.weather.gov.hk/contente.htm) were acquired. Average daily temperatures and relative humidities were 24.1 ± 4.7 °C and 79.8 ± 11.1% (Supplemental Table S1), respectively. Fig. 1 locates the seven ambient monitoring stations and participants' residences, with distances from the stations ranging 1.0–31.2 km with an average of ~14.5 km. It is assumed that the distances <20 km do not affect the estimated P-C associations (Sarnat et al., 2010). Quality assurance and quality control procedures are detailed in the Supporting Information.

2.1.3. Filter analyses

Triplicate Teflon-membrane filter weights (±3 μg) were determined using a microbalance (readability of 1 μg, Sartorius AG, Model ME 5-0 CE, Goettingen, Germany) in a temperature (20–25 °C) and relativity humidity (35 ± 5%) controlled environment. Averages of the triplicate post- and pre-weights were used to calculate mass concentrations.

Teflon-membrane filter samples were transmitted to the Desert Research Institute laboratories (DRI, Reno, NV, USA) in a temperature-controlled package (<4 °C) for elemental analysis (Watson et al., 1999) by an Energy Dispersive X-Ray Fluorescence analyzer (ED-XRF, Epsilon 5, PANalytical Company, Almelo, The Netherlands) for 51 elements (sodium to lead) (Chow and Watson, 2012). 19 elements (i.e., 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]) returned concentrations exceeding minimum detection limits (MDL) for >50% of the samples, and these elements are included in data analysis. Field blanks were analyzed following the same procedure. MDLs were within the range of 0.5 to 33 ng/m^3. The 19 elements were detectable (i.e., >MDLs) for >85% of the samples with the exception of Mg, Cr, and As (54–75% detectable).

2.2. Estimation of personal exposure to $PM_{2.5}$ of ambient origin (E_a)

Total personal $PM_{2.5}$ exposure is the sum of E_a and E_{na} (Wallace and Williams, 2005; Wilson and Brauer, 2006). E_a consists of infiltrated $PM_{2.5}$ when subjects remain indoors and direct exposure while subjects are outdoors. E_{na} accounts for exposure due to indoor sources and personal activities while subjects stay indoors and outdoors (Noullett et al., 2010; Wilson and Brauer, 2006; Wilson et al., 2000). Personal exposures and ambient concentrations can be measured directly while E_a and E_{na} can only be estimated (Wilson and Brauer, 2006). The equations are given by the following expression:

$$P = E_a + E_{na} \tag{1}$$

$$P = F_{pex}C + E_{na} \tag{2}$$

$$F_{pex} = S_{P_{ij}}/S_{C_j} \tag{3}$$

where the exposure factor (F_{pex}), i.e., personal-to-ambient sulfur ratio, estimates the contribution of ambient particles to total personal exposures; S_{Pij} represents personal exposure to sulfur for subject i on jth day; and S_{Cj} represents the ambient sulfur concentration measured at the fixed monitoring station on jth day (Ebelt et al., 2005; Wilson et al., 2000).

2.3. Statistical analysis

A paired sample t-test was applied to compare the mass and elemental concentration differences between personal exposures and the corresponding ambient concentrations. Spearman's r_s was used where the data were not normally distributed; otherwise, Pearson's correlations (r) was applied. Linear regression analysis was used to analyze the strength of P-C associations. A p-value of <0.05 is considered statistically significant.

The uniformity between ambient and personal exposures was assessed using the coefficients of divergence (COD) (Krudysz et al., 2008). Two datasets are more similar (i.e., negligible differences in absolute concentrations) when COD approaches zero (e.g., <0.20) and more different when COD values approach one. COD is calculated as follows:

$$\mathrm{COD_{jk}} = \sqrt{\frac{1}{n}\sum_{i=1}^{n}\left(\frac{\mathrm{X_{ij}}-\mathrm{X_{ik}}}{\mathrm{X_{ij}}+\mathrm{X_{ik}}}\right)^2} \tag{4}$$

where X_{ij} and X_{ik} represent ith observation of chemical component X at the central monitoring stations (or for subjects) throughout the sampling period; j and k represent concentrations from the two different sampling stations that are compared, and n is the observation number.

A mixed-effects model was used to analyze calibration coefficients using ambient concentrations as fixed-effects variables and study subjects as random variables to account for between-individual variance. The calibration coefficients (β) were estimated as the fixed regression coefficients:

$$Y_{ij} = \mu_Y + \beta_1 C_{ij} + \beta_2 Season_{ij} + \beta_3 Met_{ij} + b_i + \varepsilon_{ij} \quad (5)$$

where Y_{ij} represents the "true exposures" (either E_a or P), and C_{ij} represents surrogate exposures (i.e., ambient $PM_{2.5}$ concentrations) for *i*th subject on *j*th day (Zeger et al., 2000). Calibration coefficients equal to unity suggested no bias, while less than one suggested an attenuated estimate (Kioumourtzoglou et al., 2014). The mixed-effects model (Eq. (5)) was implemented by controlling seasonality ($Season_{ij}$) and meteorological conditions (Met_{ij}) with the assumption that the random effects (b_i and ε_{ij}) are mutually independent with a mean of zero as well as within-individual (σ^2_b) and between-individual (σ^2_w) variance. The mixed-effects model was applied in the statistical environment R 3.4.1 (The R Project for Statistical Computing, 2018: http://www.r-project.org). The marginal R^2 statistic was used to measure the overall predictive ability; the semi-partial R^2 statistic (R^2_β) was calculated for each variable (Jaeger et al., 2016).

3. Results

3.1. Activity profiles

A summary of subjects' activity patterns is presented in Table 1. 229 (94.6%) activity diaries were considered valid (i.e., complete activity logs corresponding to 1440 min sampling durations). On average, subjects spent 84.1 ± 14.5% of each day indoors, of which 71.5 ± 19.2% were at home. Outdoor activities accounted for 11.9 ± 12.7% of the time with the remaining time spent in transportation (4.0 ± 4.3%), indoors at work/school (4.8 ± 10.9%), or inside other buildings (4.3 ± 10.5%); negligible amount of time (~2.0%) was spent cleaning. Higher standard deviations associated with averages imply large variations in daily activities. Subject-specific activity patterns are summarized in Table S3. For Hong Kong residences, Chau et al. (2002) reported that people spent 86% of the time indoors, 3–7% in transit, and 3–7% outdoors. A recent 48-subject panel study also found that residents spent 69.4–73.6% of their time indoors at home, 4.0–5.9% in transit, and 5.1–5.3% outdoors. Indoor cooking/dining and cleaning activities constituted 1.7–3.3% and 4.7–7.5% of the day, respectively. These personal activities (including time indoors, in transit, cooking, and cleaning) were positively associated with personal $PM_{2.5}$ exposures (Chen et al., 2018).

Table 1
Summary of study subjects and information from the questionnaire and time-activity diaries.

Personal characteristics[a]	
Sampling date	April 25, 2014–June 7, 2015
Study subjects (N)	13
Female	6
Male	7
Median age, years (range)	37 (19–57)
Smokers (Yes/No, %)	No (100%)
Environmental tobacco smoke (ETS)[b]	
Rarely	10 (76.9%)
Outdoors (e.g., street)	3 (23.1%)
Number of valid personal measurements (%)[c]	242 (97.6%)
Personal activity diaries (%)[d]	229 (94.6%)
Time-activities data from diaries (n = 229)	**Mean ± SD[e]**
Indoors, total	84.1 ± 14.5%
Indoors, at home	71.5 ± 19.2%
Sleeping	36.7 ± 7.2%
Sitting	24.8 ± 16.8%
Cooking/Dining	8.0 ± 6.1%
Cleaning activity	2.0 ± 3.5%
Indoors, but not in a residential home (e.g., at chool, office)	4.8 ± 10.9%
Inside other buildings (e.g., canteen, shopping mall, gymnasium)	4.3 ± 10.5%
Transportation	4.0 ± 4.3%
Bus/mini-bus	2.5 ± 3.9%
Metro	1.5 ± 3.0%
Outdoors (e.g., walking outside)	11.9 ± 12.7%

[a] Reported values calculated from daily self-reporting individual activities; data were collected over 229 days from 13 subjects; mean values are weighted averages based on individual 15-min intervals summed over the entire sampling period.
[b] Information from questionnaire, no available detailed data (e.g., frequency, duration) from daily activity diaries.
[c] Percent of samples collected.
[d] Percent of valid data.
[e] SD refers to standard deviation.

3.2. Characteristics of personal $PM_{2.5}$ exposures

Fig. 2 illustrates the temporal variations in subject-specific $PM_{2.5}$ exposure. Significant seasonal differences ($p < 0.001$) were found with the highest exposure levels in winter and lowest in summer. Daily personal $PM_{2.5}$ exposures ranging from 11.6 to 80.8 μg/m^3 with an average of 33.7 ± 14.8 μg/m^3. Table 2 shows that annual average personal $PM_{2.5}$ exposures ranged from 22.3 ± 11.2 μg/m^3 to 41.7 ± 18.9 μg/m^3 by subject. The 30-day moving average shows similar peaks and valleys between personal exposure and ambient $PM_{2.5}$ with consistently higher concentrations found in personal samples (Fig. 2).

3.3. Association of personal exposures and ambient concentrations

3.3.1. $PM_{2.5}$ mass concentrations

No significant mass differences (2.2 μg/m^3, 95% Confidence Interval: 1.7–6.1 μg/m^3) were observed between Partisol sampler and PEMs ($p = 0.24$) (Fig. S1). High Spearman's correlations ($0.90 < r_s < 0.99$; $p < 0.01$) and low COD values (ranging 0.10–0.36) were found among the seven monitoring stations (Table S2 and Fig. S2), indicating a homogenous distribution in outdoor $PM_{2.5}$ concentrations across the study area.

As shown in Table 2, personal exposures were significantly higher ($p < 0.05$) compared to ambient $PM_{2.5}$ during all seasons with seasonal personal-to-ambient (P/C) $PM_{2.5}$ ratios greater than one. Only two subjects (i.e., IDs 101 and 113) had lower personal exposure levels with average and median P/C ratios ranging 0.86–0.94.

Table 3 summarizes the regression statistics of personal exposure vs. ambient $PM_{2.5}$ concentration by season (across all subjects) and by subject (across all seasons for each subject). Moderate to strong Pearson's correlations were reported by season ($0.44 < r < 0.79$; $p < 0.01$) and by subject ($0.58 < r < 0.96$). The average intercept was 9.3 ± 5.2 μg/m^3 (ranging 0.4–22 μg/m^3) with ~50% of the samples having intercepts >6.1 μg/m^3 (Supporting Information), indicating a considerable portion of fine particles were generated indoors and/or due to personal activities.

3.3.2. $PM_{2.5}$ elements

Summary statistics for the elemental composition of personal $PM_{2.5}$ along with P/C ratios are shown in Table 4. The largest mass difference was found for Na (1529 ng/m^3; $p < 0.001$), with large differences also found for Cl (57 ng/m^3; $p < 0.001$), Ca (65 ng/m^3; $p < 0.001$), Ti (3 ng/m^3; $p < 0.001$), and Fe (52 ng/m^3; $p < 0.001$). Sulfur was the most abundant element measured in ambient $PM_{2.5}$ (2921 ng/m^3), accounting for 10.9% of the $PM_{2.5}$ mass (Table S1). Strong correlations ($0.92 < r < 0.96$; $p < 0.01$) were found between sulfur and sulfate (with sulfate to sulfur ratios of 3.2–3.8) for both summer and winter (Fig. S3). Ambient concentrations were higher than personal exposures for Al, S, V, Mn, Ni, Cu, Zn, As, and Pb ($p < 0.05$), with mean and median P/C ratios in the range of 0.60–0.90 (Table 4).

Fig. 3a shows higher P-C Spearman's correlation coefficients (r_s) for sulfur across subjects ($0.82 < r_s < 0.98$; $p < 0.01$). Strong correlations were also found for Si, Zn, Br, and Pb with median r_s ranging

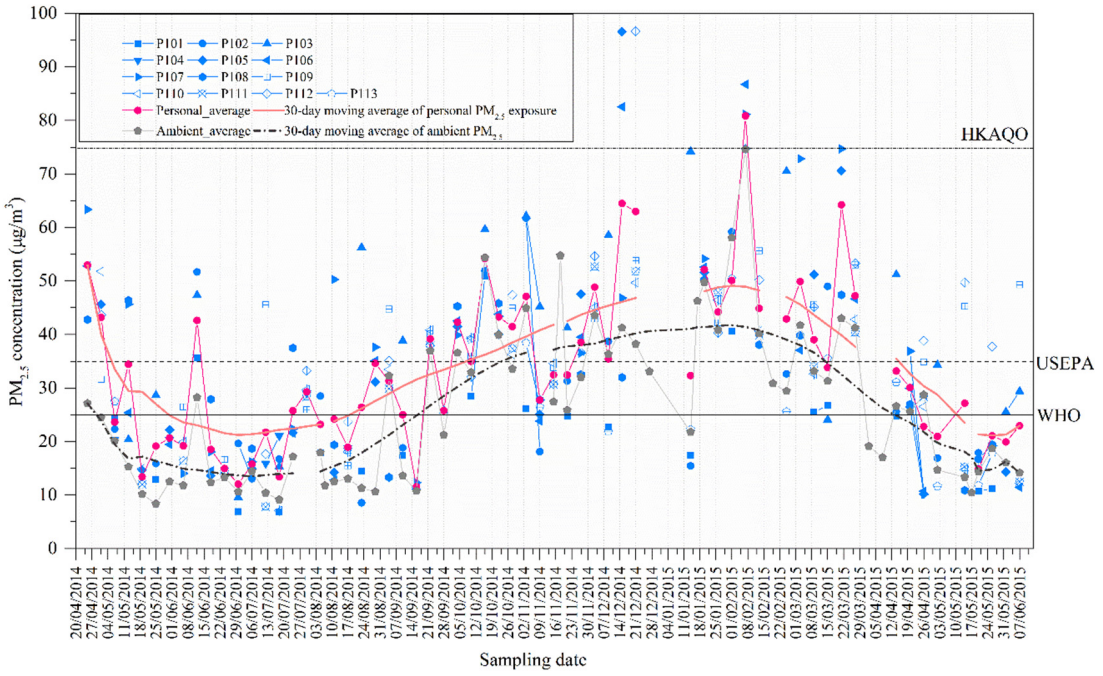

Fig. 2. Subject-specific $PM_{2.5}$ exposures (μg/m^3) and time series of daily average ambient and personal $PM_{2.5}$ throughout the study period.

0.86–0.90 ($p < 0.05$), and weak or no associations for Na, Mg, and Cr. Moderate P-C correlations were found for Ca (median $r_s = 0.58$; $p < 0.05$), Fe (median $r_s = 0.59$; $p < 0.05$), and Ti (median $r_s = 0.64$, $p < 0.05$). Fig. 3b shows higher COD values (0.20 < COD < 0.50) for soil dust (e.g., Al, Si, K, Ca, Fe, Ti, and Zn) and other trace elements (e.g., V, Ni, and Cu). A COD value of >0.20 was adapted as a threshold to illustrate the disparity between ambient concentrations and personal exposures (Kim and Hopke, 2008). Heterogeneous distributions (COD >0.50) between ambient concentrations and personal exposures were also found for Na, Cl, Mg, and Cr. The lowest COD values (ranging 0.09–0.20) were found for sulfur (Fig. S2), indicating its uniform distribution across the study area.

3.4. Characterization of personal exposure to $PM_{2.5}$ of ambient origin

Fig. 4 illustrates the linear regression of the pooled personal exposures and ambient concentrations for sulfur ($N_p = 230$). Regression slope provides a general estimate of the average ambient exposure

Table 2
Statistical summary of personal $PM_{2.5}$ exposure and personal-to-ambient $PM_{2.5}$ ratios by season and by subject, along with mass differences between personal exposures (variable 1) and ambient concentrations (variable 2)[e].

		Personal $PM_{2.5}$ exposure (μg/m^3)					Personal-to-ambient (P/C) ratio (no unit)					Mass difference (μg/m^3)	*p*-value[f]
		Mean ± SD[a]	Median	Min-Max	Q1-Q3[b]	N[c]	Mean ± SD[a]	Median	Min-Max	Q1-Q3[b]	Np[d]	(Mean ± SD[a])	
Season[g]	Spring	32.1 ± 15.8	28.5	10.8–74.7	19.4–45.2	76	1.44 ± 0.66	1.22	0.60–3.73	1.03–1.63	76	8.4 ± 11.5	< **0.001**
	Summer	22.4 ± 11.8	18.1	6.8–56.2	14.3–28.5	65	1.62 ± 0.92	1.29	0.65–4.99	1.07–1.82	60	8.0 ± 10.9	< **0.001**
	Autumn	35.3 ± 11.9	36.6	11.1–62.2	28.2–42.0	59	1.15 ± 0.31	1.11	0.41–2.86	1.03–1.23	52	3.9 ± 7.5	**0.001**
	Winter	50.2 ± 19.9	49.9	15.5–96.6	39.9–55.4	42	1.23 ± 0.55	1.06	0.60–3.41	0.99–1.25	42	7.9 ± 17.7	**0.006**
Subject ID	101	22.3 ± 11.2	23.5	6.8–50.9	14.1–26.3	20	0.92 ± 0.27	0.86	0.58–1.54	0.74–1.05	20	(-)3.6 ± 7.0	**0.03**
	102	29.2 ± 16.4	22.3	8.5–61.7	16.9–38.7	21	1.28 ± 0.37	1.18	0.71–1.89	1.01–1.57	20	5.2 ± 7.5	**0.006**
	103	41.7 ± 18.9	41.3	9.5–74.2	27.1–57.4	19	2.06 ± 1.06	1.68	0.77–4.99	1.44–2.38	18	**18.4 ± 15.8**	< **0.001**
	104	23.7 ± 7.3	20.7	15.9–35.8	20.3–27.0	6	1.55 ± 0.49	1.53	1.01–2.30	1.27–1.63	5	6.6 ± 4.3	**0.03**
	105	36.4 ± 23.7	28.8	11.1–96.5	14.8–48.5	20	1.41 ± 0.53	1.14	0.89–2.93	1.03–1.59	19	**9.8 ± 14.1**	**0.007**
	106	34.4 ± 21.5	29.9	11.4–86.7	16.7–43.8	21	1.35 ± 0.56	1.16	0.81–3.31	1.09–1.33	20	7.8 ± 11.0	**0.005**
	107	40.7 ± 20.9	38.7	12.3–81.1	21.6–51.2	20	1.67 ± 0.90	1.19	1.03–4.02	1.11–1.74	19	**13.2 ± 13.7**	**0.001**
	108	31.0 ± 13.0	32.0	10.9–50.2	19.4–42.8	21	1.32 ± 0.71	1.08	0.41–3.71	1.00–1.36	20	4.3 ± 11.1	**0.10**
	109	38.0 ± 12.4	42.2	15.1–55.6	31.6–45.5	21	1.66 ± 0.96	1.29	0.99–4.38	1.16–1.39	20	**10.4 ± 11.0**	< **0.001**
	110	31.8 ± 13.4	34.0	7.1–51.8	19.4–41.3	20	1.18 ± 0.31	1.11	0.78–2.12	1.02–1.28	19	4.3 ± 6.7	**0.01**
	111	29.6 ± 13.5	29.9	7.8–52.6	16.4–38.9	21	1.11 ± 0.18	1.12	0.76–1.42	0.99–1.19	20	2.9 ± 4.2	**0.006**
	112	39.7 ± 18.3	38.3	12.0–96.6	31.7–47.9	20	1.52 ± 0.67	1.29	0.97–3.73	1.10–1.74	19	**12.0 ± 14.4**	**0.002**
	113	27.4 ± 11.5	26.0	11.6–50.4	20.8–35.8	12	0.94 ± 0.18	0.87	0.60–1.17	0.84–1.12	11	*(-)2.1 ± 6.0*	0.28
Total		33.4 ± 17.3	31.8	6.8–96.6	18.5–45.1	242	1.38 ± 0.69	1.16	0.41–4.99	1.02–1.48	230	7.2 ± 12.1	< **0.001**

[a] SD refers to standard deviation.
[b] Q1: 25th percentile; Q3: 75th percentile.
[c] N refers to the number of valid data.
[d] N_p refers to number of personal-ambient data pairs compared.
[e] Ambient $PM_{2.5}$ concentrations obtained from seven Environmental Protection Department (EPD) monitoring stations in Hong Kong.
[f] Bolded value indicated paired variables were statistically significant at the 0.05 level. *Italics* indicated mean <0.
[g] Spring (April 25th-May 31st, 2014 and March 3rd-May 26th, 2015); Summer (June 6th-August 29th, 2014 and June 1st-June 7th, 2015); Autumn (September 4th-November 27th, 2014); Winter (December 3rd, 2014-February 25th, 2015).

Table 3
Regression statistics for personal exposures and ambient $PM_{2.5}$ concentrations by season and by subject.

		Slope	Intercept, μg/m^3	Pearson's r[a]	*p*-Value[b]	N_p
Season	Spring	1.05 ± 0.13	7.3 ± 3.3	0.69	<**0.001**	76
	Summer	1.18 ± 0.32	5.6 ± 4.6	0.44	<**0.001**	60
	Autumn	0.90 ± 0.10	7.3 ± 3.4	0.79	<**0.001**	52
	Winter	0.76 ± 0.22	18.2 ± 9.6	0.48	**0.001**	42
	Mean	0.97 ± 0.18	9.6 ± 5.8	0.60		230
Subject ID	101	0.68 ± 0.09	4.6 ± 2.6	0.88	<**0.001**	20
	102	1.02 ± 0.12	4.8 ± 3.4	0.89	<**0.001**	20
	103	0.88 ± 0.31	21.2 ± 8.1	0.58	**0.01**	18
	104	0.81 ± 0.28	9.7 ± 5.0	0.85	0.07	5
	105	1.15 ± 0.19	5.9 ± 6.2	0.82	<**0.001**	19
	106	1.11 ± 0.15	4.7 ± 4.7	0.87	<**0.001**	20
	107	0.94 ± 0.19	14.8 ± 6.3	0.76	<**0.001**	19
	108	0.66 ± 0.18	13.5 ± 5.4	0.65	**0.002**	20
	109	0.61 ± 0.20	22.0 ± 5.9	0.58	**0.007**	20
	110	1.00 ± 0.14	4.3 ± 4.1	0.87	<**0.001**	19
	111	1.09 ± 0.08	0.4 ± 2.3	0.96	<**0.001**	20
	112	1.04 ± 0.30	10.9 ± 9.1	0.64	**0.003**	19
	113	0.81 ± 0.13	3.7 ± 4.3	0.90	<**0.001**	11
	Mean	0.92 ± 0.18	9.3 ± 5.2	0.79		230

Notes: N_p denotes number of personal-ambient data pairs compared.
[a] Spearman's correlation coefficients were estimated when data pairs <30.
[b] Bolded value indicated paired variables were statistically significant at the 0.05 level.

factor (F_{pex}) of 0.73 ± 0.02, indicating that personal exposure to sulfur is 73% of the ambient levels and that ambient sulfur can explain over 80% of the variations in personal sulfur exposure ($R^2 = 0.82$). Subject- and season-specific linear regression analyses between personal and ambient sulfur concentrations are summarized in Table 5. Strong P-C sulfur correlations were found by subject ($0.81 < r < 0.98$; $p < 0.01$) and by season ($0.76 < r < 0.88$; $p < 0.01$).

Fig. 5 reports the contribution of ambient $PM_{2.5}$ to personal exposure for each subject and by season. Subject-specific E_a (ranging 12.6–25.1 μg/m^3; coefficient of variance, CV = 15.4%) was higher than E_{na} (ranging 4.0 to 24.3 μg/m^3, CV = 50.1%), but with a lower CV. For the pooled estimated variables (Fig. 6), total personal exposure showed strong correlations both with E_a ($r = 0.75$; $p < 0.05$) and E_{na} ($r = 0.73$; $p < 0.05$).

Longitudinal regressions of E_a vs. C for each subject are shown in Table 6. Correlations range 0.82–0.99 with an average of 0.94 by subject and 0.80–0.95 by season with an average of 0.89 (Table 6). Table 7 presents the results of mixed-effects regressions for E_a and personal $PM_{2.5}$ exposure as compared to ambient $PM_{2.5}$. Personal $PM_{2.5}$ exposure calibration coefficients of 0.87 (95% CI, 0.70–1.04, $p = 0.002$) are higher

Table 4
Statistical summary of $PM_{2.5}$ elemental concentrations for personal samples, personal to ambient elements ratios and differences in average $PM_{2.5}$ elemental concentrations between personal (variable 1) and ambient (variable 2) samples[g].

	Elements in personal $PM_{2.5}$ (ng/m^3)							P/C ratio (no unit)					Mass difference (ng/m^3) (Mean ± SD[a])	*p*-Value[h]
	Mean ± SD[a]	Median	Min-Max	Q1-Q3[b]	MDLs	N[c]	>MDL%[d]	Mean ± SD[a]	Median	Q1-Q3[b]	95 CI%[e]	N_p[f]		
Na	3037 ± 1901	2489	103–9374	1661–3848	33	244	99.2	6.91 ± 7.93	3.43	0.61–1.17	5.88–7.95	224	1529 ± 2380	<**0.001**
Mg	128 ± 86	112	13–435	57–172	1	174	70.7	1.42 ± 2.47	0.80	0.79–1.03	1.04–1.81	159	*(−31) ± 121*	**0.001**
Al	132 ± 99	124	7–619	44–166	5	232	94.3	0.92 ± 0.59	0.75	1.02–3.43	0.84–0.99	216	*(−25) ± 78*	<**0.001**
Si	256 ± 287	176	4–2412	69–328	3	233	94.7	1.14 ± 1.53	0.80	0.77–1.14	0.93–1.34	218	*(−17) ± 212*	0.25
S	2511 ± 1279	2317	85–6324	1493–3488	2	246	100.0	0.93 ± 0.26	0.90	1.12–1.94	0.89–0.96	230	*(−346) ± 710*	<**0.001**
Cl	177 ± 259	73	6–1633	31–201	5	243	98.8	3.55 ± 7.17	1.72	0.96–1.57	2.63–4.48	229	57 ± 210	<**0.001**
K	284 ± 226	238	21–1303	113–378	3	246	100.0	1.27 ± 1.91	0.91	0.42–0.88	1.02–1.51	231	*(−0.1) ± 164*	0.99
Ca	161 ± 143	128	17–1297	84–173	2	245	99.6	2.25 ± 5.46	1.39	0.58–1.98	1.55–2.96	231	65 ± 132	<**0.001**
Ti	13 ± 10	11	1–64	6–17	1	237	96.3	1.54 ± 1.26	1.23	0.57–1.06	1.37–1.71	219	3 ± 7	<**0.001**
V	16 ± 15	11	1–94	6–19	1	238	96.7	0.72 ± 0.50	0.63	0.80–1.52	0.65–0.79	224	*(−7) ± 12*	<**0.001**
Cr	4 ± 4	3	1–35	2–4	0.9	185	75.2	1.97 ± 2.98	0.98	0.52–1.15	1.52–2.41	170	*1 ± 5*	**0.003**
Mn	11 ± 9	10	1–56	4–16	0.8	225	91.5	0.90 ± 0.63	0.80	0.42–0.83	0.82–0.99	211	*(−2) ± 6*	<**0.001**
Fe	232 ± 180	185	8–1162	113–305	0.7	246	100.0	1.41 ± 1.46	1.08	0.65–1.03	1.22–1.60	231	52 ± 154	<**0.001**
Ni	6 ± 5	4	0–25	2–8	0.5	237	96.3	0.91 ± 0.55	0.79	0.42–0.78	0.84–0.98	223	*(−0.7) ± 3*	**0.004**
Cu	15 ± 20	11	1–250	5–18	0.5	241	98.0	0.74 ± 0.64	0.62	0.71–1.14	0.66–0.83	226	*(−4) ± 17*	<**0.001**
Zn	106 ± 133	68	1–1366	24–144	0.5	246	100.0	1.29 ± 3.83	0.81	0.55–0.90	0.80–1.79	231	*(−4) ± 121*	0.60
As	3 ± 2	3	1–13	2–4	0.8	132	53.7	0.65 ± 0.43	0.60	0.42–0.78	0.57–0.74	103	*(−2) ± 3*	<**0.001**
Br	11 ± 11	9	1–88	3–16	0.5	244	99.2	1.04 ± 0.81	0.87	0.71–1.14	0.94–1.15	230	*(−0.3)* ± 6	0.47
Pb	25 ± 36	19	1–441	5–35	0.5	208	84.6	0.75 ± 0.33	0.71	0.55–0.90	0.70–0.79	194	*(−6) ± 34*	**0.02**

[a] SD refers to standard deviation.
[b] Q1: 25th percentile; Q3: 75th percentile.
[c] N refers to the number of valid data, and concentrations below the minimum detection limits (MDLs) were removed.
[d] MDLs refers to method detection limits, which is three times of the standard deviation of average laboratory blanks by x-ray fluorescence (Watson et al., 1999).
[e] 95% confidential interval.
[f] N_p refers to number of personal-ambient data pairs compared.
[g] Ambient elemental concentrations obtained from seven EPD air quality monitoring stations in Hong Kong.
[h] Bolded value indicated paired variables were statistically significant at a significance level of 0.05 for a two-sided test. *Italics* indicated mean <0.

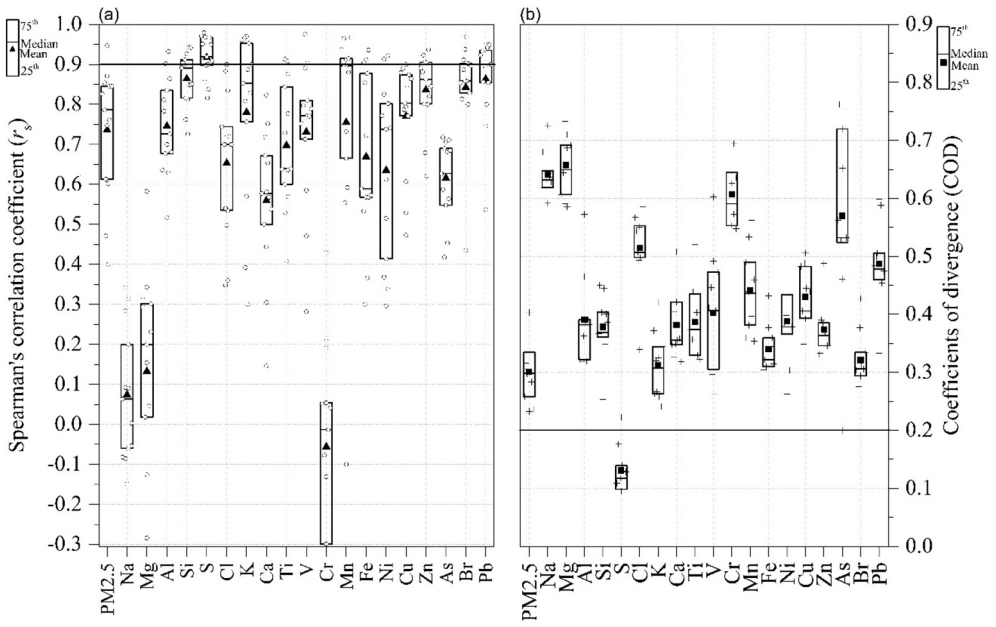

Fig. 3. Distribution of (a) subject-specific Spearman's correlation coefficients (r_s) and (b) coefficients of divergence for personal-to-ambient $PM_{2.5}$ and elements ratios. COD values >0.20 (solid line) imply similarities. Boxplots represent 75th percentile, median, and 25th percentile of all data.

than 0.78 for E_a (95% CI: 0.72–0.84, $p < 0.001$), when adjusted for seasonality and meteorological conditions.

4. Discussion

Daily average personal $PM_{2.5}$ exposure exceeded the World Health Organization (WHO) 24-h air quality guideline of 25 μg/m^3 for 67.2% of the sampling days, mostly during winter. These results agree with previous studies that shown higher personal $PM_{2.5}$ levels in Hong Kong than those in many North American and European cities (ranging 12.9–25.4 μg/m^3) (Kim et al., 2005; Kioumourtzoglou et al., 2014; Meng et al., 2012; Noullett et al., 2010; Wallace and Williams, 2005; Williams et al., 2000). However, average personal $PM_{2.5}$ exposures of 33.4 ± 17.3 μg/m^3 (ranging 22.4–50.2 μg/m^3) in this study are ~50% lower than those in Chinese cities (ranging 45.4–126.8 μg/m^3) (e.g., Guangzhou, Shanghai, Tianjin, Beijing) (Baccarelli et al., 2014; Hu et al., 2018; Jahn et al., 2013; Lei et al., 2016) and in New Delhi, India (ranging 53.9–489.2 μg/m^3) (Pant et al., 2017).

Personal $PM_{2.5}$ exposures exceeding the corresponding ambient (or outdoor) $PM_{2.5}$ concentrations were also reported in past studies (Fan et al., 2017; Hsu et al., 2012; Hu et al., 2018; Jahn et al., 2013; Kioumourtzoglou et al., 2014; Meng et al., 2012). Table 2 shows that four of the 13 (30.8%) subjects (e.g., IDs: 103, 107, 109, and 112) reported annual average exposures of 38.0–41.7 μg/m^3, with P-C mass differences of 10.8–18.6 μg/m^3 ($p < 0.05$). Moderate to strong P-C correlations ($0.58 < r < 0.76$; $p < 0.01$) and higher $PM_{2.5}$ P/C ratios (1.5–2.1) were also found. Higher personal to ambient mass differences may be due to the between-individual variance related to their daily

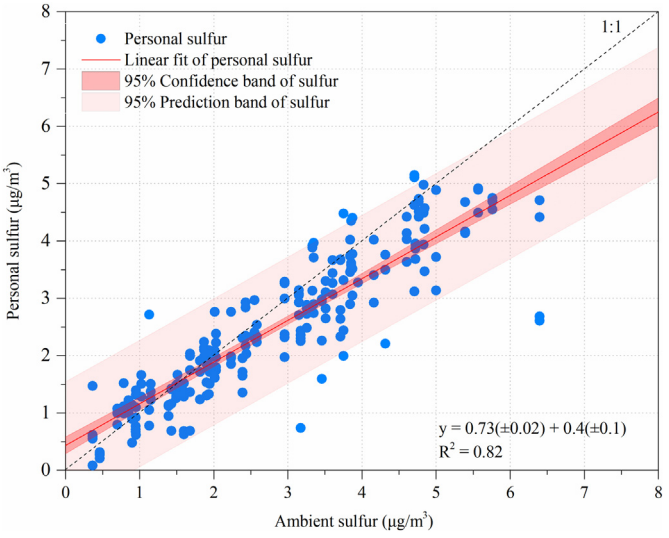

Fig. 4. Association of ambient and personal exposure to sulfur throughout the study period ($N_p = 230$).

Table 5
Regression statistics for personal exposure and ambient sulfur concentrations by season and by subject.

		Slope	Intercept, μg/m^3	Pearson's r[b]	*p*-Value[c]	N_p
Season	Spring	0.77 ± 0.05	0.3 ± 0.1	0.88	<**0.001**	76
	Summer	0.83 ± 0.06	0.3 ± 0.1	0.88	<**0.001**	60
	Autumn	0.67 ± 0.05	0.5 ± 0.3	0.88	<**0.001**	52
	Winter	0.77 ± 0.10	0.5 ± 0.4	0.76	<**0.001**	42
	Mean	0.76 ± 0.07	0.4 ± 0.1	0.85		
Subject ID	101	0.60 ± 0.08	0.4 ± 0.3	0.87	<**0.001**	20
	102	0.72 ± 0.06	0.4 ± 0.2	0.95	<**0.001**	20
	103	0.44 ± 0.08	1.0 ± 0.3	0.81	<**0.001**	18
	104	0.78 ± 0.12	0.3 ± 0.3	0.97	**0.007**	5
	105	0.89 ± 0.05	0.1 ± 0.2	0.98	<**0.001**	19
	106	0.90 ± 0.05	0.1 ± 0.2	0.97	<**0.001**	20
	107	0.77 ± 0.06	0.5 ± 0.2	0.95	<**0.001**	19
	108	0.75 ± 0.09	0.3 ± 0.3	0.90	<**0.001**	20
	109	0.66 ± 0.07	0.6 ± 0.2	0.92	<**0.001**	20
	110	0.82 ± 0.06	0.3 ± 0.2	0.95	<**0.001**	19
	111	0.89 ± 0.06	0.2 ± 0.2	0.96	<**0.001**	20
	112	0.86 ± 0.08	0.2 ± 0.3	0.93	<**0.001**	19
	113	0.65 ± 0.14	0.3 ± 0.5	0.83	**0.002**	11
	Mean	0.75 ± 0.07	0.4 ± 0.2	0.92		
Total[a]		0.73 ± 0.03	0.3 ± 0.1		<**0.001**	230

Notes: N_p: number of personal-ambient data pairs compared.
[a] Results are from mixed-effects regression analysis, which account for the repeated measurements within subject ($\sigma^2_b = 0.003$ and $\sigma^2_w = 0.08$) while controlling for seasonality and meteorological conditions (e.g., T, RH).
[b] Spearman's correlation coefficients (r_s) were estimated when data pairs <30.
[c] Bolded value indicated paired variables were statistically significant at the 0.05 level.

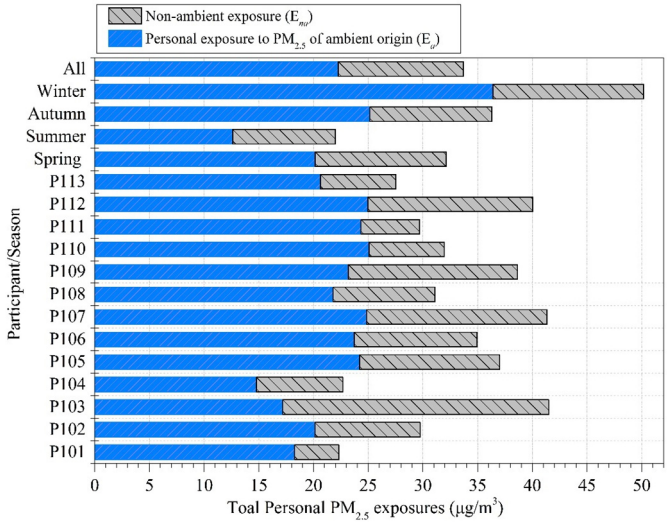

Fig. 5. Estimation of personal exposure to $PM_{2.5}$ of ambient origin (E_a) and non-ambient exposure (E_{na}) by subject and by season.

activities or lifestyles (Table S3). Past findings have indicated that subjects who were more active may have higher and more variable exposures than the corresponding ambient concentrations (Baccarelli et al., 2014). For adult subjects in Guangzhou, Jahn et al. (2013) reported that five out of seven districts showed higher personal $PM_{2.5}$ exposures than ambient concentrations, attributing the increments to indoor sources and personal activities. Health estimates in an epidemiologic analysis would be underestimated with weak P-C correlations. Fan et al. (2018) reported higher personal $PM_{2.5}$ exposures with moderate P-C correlation ($r_s = 0.52$; $p < 0.05$) for healthy residents in Hong Kong. They found that an interquartile change (16.4 μg/m^3) in personal $PM_{2.5}$ exposure was linked to a 12.8% (95 CI%, 5.5–20.7%) increase in FeNO (i.e., fractional nitric oxide concentration in exhaled breath, a biomarker of airway inflammation); no positive association was shown for ambient $PM_{2.5}$ concentrations.

This study provides evidence that longitudinal P-C correlations ($r = 0.88$) (data not shown) could be a better indicator than those of cross-sectional correlations ($r = 0.60$, Table 3). Personal exposure across a subpopulation leads to improved P-C correlations than individual exposures (Jahn et al., 2013; Rhomberg et al., 2011; Zeger et al., 2000). Since 11 of the 13 subjects were sampled at least 20 times, within-individual P-C $PM_{2.5}$ correlations are higher (Avery et al., 2010). The results are also consistent with decreased personal-ambient mass differences being associated with increased P-C correlations (Adgate et al., 2003; Avery et al., 2010).

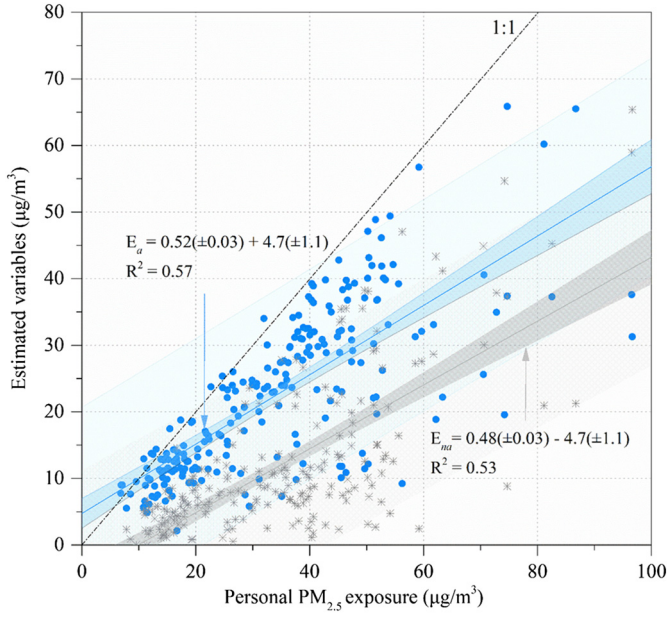

Fig. 6. Scatter plot and regression statistics with 95% confidence interval showing the relationship between estimate variables (E_a, E_{na}) with total personal exposures.

Table 3 shows lower P-C $PM_{2.5}$ correlations during summer ($r = 0.44$, $p < 0.01$) and winter ($r = 0.48$, $p < 0.01$) compared to other seasons, which could result from reduced infiltration rates due to the closed windows for cooling in summer and for thermal comfort in winter. Sarnat et al. (2000) and Sarnat et al. (2006) suggested that season is an essential factor in determining P-C associations. Chen et al. (2018) also confirmed that season was a factor affecting the strength of personal-ambient $PM_{2.5}$ associations.

COD analyses reveal dissimilarities between personal exposures and ambient $PM_{2.5}$ concentrations for mass (ranging 0.15–0.41 with median COD = 0.23) and most elements (median COD ranging 0.24–0.62). The personal $PM_{2.5}$ samples were enriched (P/C ratios >1) in Ca and Ti (i.e., resuspension particles), as well as Fe and Cr (e.g., traffic-related particles), consistent with daily activity patterns affecting total personal exposures (Chen et al., 2018; Krall et al., 2018). Larson et al. (2004) and Hsu et al. (2012) reported high associations between personal activities (e.g., time indoors) and crustal particles (e.g., Ca). Average P/C ratios >1 for Zn (Table 4) may be impacted by both indoor (e.g., cleaning products) and outdoor (e.g., traffic) sources. P/C data with average and median P/C ratios <1 (e.g., S, Mn, V, Ni, and As) were less influenced by indoor sources or personal activities (Hsu et al., 2012).

Greater spatial variability was found for most of the analyzed elements (as compared to $PM_{2.5}$ mass and sulfur) with higher COD values and weak P-C correlations, consistent with higher exposure measurement errors (Dionisio et al., 2016). To quantify the effects of exposure error on health risk estimates, Dionisio et al. (2016) showed that spatial errors in epidemiological models have the potential to introduce 10–40% biases in relative risk estimates.

The spatial distribution for sulfur was more uniform than for other elements with P-C correlations >0.90 ($p < 0.01$) and COD values <0.20, consistent with previous studies (Brokamp et al., 2015; Hsu et al., 2012; Wallace and Williams, 2005). Not much variation by season was found for F_{pex} (ranging 0.67–0.83). Differences in subject-specific F_{pex} (ranging 0.44–0.90) were mostly due to subjects' daily activity patterns. Replacing P/C sulfur ratios with slopes from the personal vs. ambient sulfur regressions by subject and by season improved F_{pex} (0.73–0.76, averaging 0.73 ± 0.03). These are comparable to previous findings (0.54–0.75) using sulfate or sulfur as an outdoor exposure marker (Allen et al., 2004; Kioumourtzoglou et al., 2014; Noullett et al., 2010; Wilson and Brauer, 2006). The adjusted F_{pex} should be considered as upper bounds because it is assumed that infiltration is proportional to ambient concentrations (Ott et al., 2000). Chen et al. (2017) found that sulfate can be used to estimate E_a for adult subjects in Guangzhou, China. In contrast, Ji et al. (2018) suggested sulfur may not be a sufficient indicator in Beijing, China; they proposed the use of iron to estimate indoor $PM_{2.5}$ of outdoor origin.

Average E_{na} (11.2 ± 5.6 μg/m^3) in Hong Kong was higher than in Vancouver, Canada (8.47 μg/m^3) (Wilson and Brauer, 2006); Prince George, Canada (5.0–6.4 μg/m^3) (Noullett et al., 2010); and in the U.S. (10.0–12.5 μg/m^3) (Schwartz et al., 2007; Williams et al., 2003). Higher E_{na} values were reported in Guangzhou (18.1 ± 29.1 μg/m^3) and in Tianjin (63–97 μg/m^3, for PM_{10}), China (Chen et al., 2017; Xu et al., 2014). As shown in Fig. 6, on average, 52% of the personal exposure is due to E_a and 48% to E_{na}. Comparable contributions and similar correlations of E_a and E_{na} with personal exposure illustrate the importance of both ambient and indoors sources and their relevance to personal activities.

Table 6
Regression statistics for personal exposure to $PM_{2.5}$ of ambient origin (E_a) vs. ambient concentration (C) by season and by subject.

		Slope	Intercept, μg/m³	Pearson's r[a]	p-Value[b]	N_p
Season	Spring	0.87 ± 0.03	*(−0.5) ± 0.8*	0.95	<**0.001**	76
	Summer	0.94 ± 0.06	*(−0.5) ± 0.8*	0.91	<**0.001**	60
	Autumn	0.67 ± 0.07	3.1 ± 2.4	0.80	<**0.001**	52
	Winter	0.88 ± 0.06	*(−0.7) ± 2.8*	0.91	<**0.001**	42
	Mean	0.84 ± 0.06		0.89		230
Subject ID	101	0.60 ± 0.07	2.7 ± 2.1	0.90	<**0.001**	20
	102	0.84 ± 0.06	*(−0.4) ± 1.6*	0.96	<**0.001**	20
	103	0.53 ± 0.09	4.8 ± 2.4	0.82	<**0.001**	18
	104	0.75 ± 0.14	2.7 ± 2.5	0.95	**0.01**	5
	105	0.91 ± 0.03	*(−0.7) ± 0.9*	0.99	<**0.001**	19
	106	0.90 ± 0.02	*(−0.8) ± 0.8*	0.99	<**0.001**	20
	107	0.85 ± 0.03	0.8 ± 1.0	0.99	<**0.001**	19
	108	0.82 ± 0.09	*0 ± 2.8*	0.90	<**0.001**	20
	109	0.79 ± 0.07	1.6 ± 2.0	0.94	<**0.001**	20
	110	0.92 ± 0.05	*(−0.4) ± 1.6*	0.97	<**0.001**	19
	111	0.94 ± 0.04	*(−0.7) ± 1.2*	0.98	<**0.001**	20
	112	0.91 ± 0.06	*(−0.4) ± 1.8*	0.96	<**0.001**	19
	113	0.75 ± 0.11	(−1.5) ± 3.7	0.91	<**0.001**	11
	Mean	0.60 ± 0.20		0.94		230

Notes: N_p denotes the number of personal-ambient data pairs compared.
[a] Spearman's correlation coefficients (r_s) were estimated when data pairs <30.
[b] Bolded value indicated paired variables were statistically significant at the 0.05 level. *Italics* indicated mean <0.

The strength of P-C and E_a-C associations vary across subjects and seasons, indicating the presence of intra- and inter-personal heterogeneity. Higher correlation coefficients were found between spatially-averaged ambient $PM_{2.5}$ with E_a (Adj. $R^2 = 0.90$; $p < 0.01$) than among individual subjects (Adj. $R^2 = 0.58$, $p < 0.01$). Sheppard et al. (2005) showed that increases in the number of central monitors is associated with decreases in measurement error. Calibration coefficient < 1 agree with those estimated with a regression model in the time-series studies by Kioumourtzoglou et al. (2014). Substantial attenuations of 0.31–0.39 were reported in other studies (Sarnat et al., 2001; Schwartz et al., 2007) using ambient $PM_{2.5}$ as the true exposure. These results refer to the proportionality coefficient discussed by Zeger et al. (2000). If there were a true health risk associated with E_a, similar results would be found for ambient $PM_{2.5}$ with lower correlation coefficients. In contrast to the Berson error, Zeger et al. (2000) indicated that the error term of average personal exposure and true ambient exposure (i.e., E_{na}) is the classical error type and has the potential to bias the estimate coefficients when E_a correlates with E_{na}. In this study, E_{na} is independent of ambient $PM_{2.5}$, as shown by very weak C-E_{na} correlations ($r = 0.09$; $p > 0.05$). Sheppard et al. (2005) contend that when ambient and non-ambient sources are independent, exposure variations due to E_{na} do not bias the effect estimates when the study targets ambient exposure effects. A lack of bias does not indicate the absence of exposure measurement error. Goldman et al. (2011) propose that measurement error reduce the statistical significance of risk ratio estimates for both error types. In this analysis, calibration coefficients <1 were found for average ambient $PM_{2.5}$, indicating the observed effect could be underestimated when ambient levels at central monitoring stations are used as exposure metrics in time-series studies.

The current analysis does not further characterize the measurement errors due to instrument imprecision and $PM_{2.5}$ spatial variability (Section 2.1.2) but utilizes the mixed-effects model to calculate exposure measurement error on C-E_a and P-C correlations. This results in some limitations: 1) the small number of subjects may limit the prediction power; 2) the Hong Kong locale may limit the generalizability to other subpopulations or cities/regions; and 3) due to the lack of health data, the bias of health risk estimates was not evaluated.

Table 7
Statistical parameters for the personal exposure to $PM_{2.5}$ of ambient origin (E_a) and total personal $PM_{2.5}$ exposure (P) with ambient $PM_{2.5}$ (C).

	Ambient $PM_{2.5}$ (C)
Personal exposure to $PM_{2.5}$ of ambient origin (E_a)	7 sites
Estimate β (95% CI)[a]	0.78 (0.72–0.84)***
p-Value	<0.001
Adj. R^2[b]	0.74 (**0.90**)
Contribution (%)[c]	82.2%
Total personal $PM_{2.5}$ exposure (P)	13 subjects ($N_p = 230$)
Estimate β (95% CI)[a]	0.87 (0.70–1.04)**
p-Value	0.002
Adj. R^2[b]	0.29 (**0.58**)
Contribution (%)[c]	50.0%

[a] Results are from mixed-effects regression analysis, which accounts for the repeated measurements within subject while controlling for seasonality and meteorological conditions (e.g., T, RH).
[b] The marginal R^2 statistic for the overall mixed-effects model are marked in bold (Adj. R^2).
[c] Denotes percentage of variance (Contribution = R^2_β / Adj. R^2 * 100%) explained by the fixed effects (e.g., C, P) in mixed-effects models.
** p-Value < 0.01.
*** p-Value < 0.001 for significant difference from 1.

5. Conclusions

Personal exposures provide more precise and representative estimates of health effects than measurements from the central monitoring stations. Consistent with previous studies, sulfur can be used as a surrogate for ambient $PM_{2.5}$, with an adjusted exposure factor (F_{pex}) of 0.73 ± 0.02. Moderate to strong P-C correlations were found ($0.58 < r < 0.96$) with higher correlations found between C and E_a ($0.82 < r < 0.99$; $p < 0.05$), as compared to E_{na} ($r = 0.09$; $p > 0.05$). Calibration coefficients were less than one, consistent with an underestimation of health risks when using ambient $PM_{2.5}$ as the surrogate for true exposures. This study also highlights the importance of E_{na} (18–59%) to personal $PM_{2.5}$ exposures. It is necessary to treat E_a and E_{na} as independent predictors of $PM_{2.5}$-related health effects. Longitudinal personal monitoring studies involving subpopulations varying in health status, occupations, activity levels, geographical locations, and other factors would be beneficial in future research studies to evaluate true exposures and expand upon our findings presented in this manuscript.

Declarations of interest

None.

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Author contributions

XCC was involved in data analysis and manuscript preparation. TW and KFH conceived and designed the study. JJC, JCC, and SCL performed the exposure assessment and chemical analyses. TW, JCC, JGW, and KFH revised the manuscript. HLY and NCL supervised the development of the study and manuscript evaluation. All authors have read and approved the final manuscript.

Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.scitotenv.2018.11.088.

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