Sources identification of personal exposure to fine particulate matter ($PM_{2.5}$) among adult residents of Hong Kong

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Abstract

Epidemiological studies provide evidence of the harmful effects of source-specific fine particulate matter on human health. Studies regarding relative contributions of multiple sources to personal exposure are limited and inconsistent. Personal fine particulate matter ($PM_{2.5}$) monitoring was conducted in 48 adult subjects (ages 18–63 years) in Hong Kong over two sampling periods between June 2014 and March 2015. We identified seven sources of personal $PM_{2.5}$ exposures using Positive Matrix Factorization (PMF). These sources included Regional pollution (associated with coal combustion and biomass burning), Secondary sulfate, Tailpipe exhaust, Secondary nitrate, Crustal/road dust, and Shipping emission sources. For personal $PM_{2.5}$ exposure, one additional source related to an individual's activity was found: Non-tailpipe emissions (characterized by Fe, Mn, Cr, Cu, Sr). A principal component analysis (PCA)/absolute principal component scores (PCA/APCS) along with linear mixed-effects models (LMM) was applied to the same dataset. Results showed highly similar factor/component profiles using PMF and PCA, with some discrepancies in the number of factors. Personal $PM_{2.5}$ constituents were stratified by season and by study group. Subsequently, PCA/APCs-LMM models were applied to estimate season- and group-specific sources contribution of personal $PM_{2.5}$ exposures, controlling for temperature and relative humidity. A mixed source contribution of secondary sulfate, secondary nitrate, and regional pollution was shown (35.1–43.6%), with no seasonal or occupational differences ($p > 0.05$). Shipping emissions were ubiquitous, contributing 6.3–8.8% of personal $PM_{2.5}$ exposure for all subjects. Tailpipe exhaust and traffic-related particles varied by season ($p < 0.01$) and subject groups ($p < 0.05$). Ambient $PM_{2.5}$ at fixed stations did not account for the contributions of personal activity related sources (e.g., non-tailpipe pollution), and personal exposure to tailpipe exhaust and regional pollution were underestimated. Caution should be taken

when using source-specific $PM_{2.5}$ as proxies for the corresponding personal exposures in epidemiological studies.

Keywords: Personal fine particles exposure; Ambient air pollution; Positive Matrix Factorization; Principal component analysis/Absolute Principal Component Scores; Mixed-effects model

1. Introduction

Epidemiological studies have linked ambient $PM_{2.5}$ (particles with aerodynamic diameter < 2.5 μm) pollution with adverse health outcomes globally (Franklin et al., 2006; Kim et al., 2015; Peng et al., 2009; Wong et al., 2015). Concern over the impacts of exposure measurement error on health risk estimates in time series studies created a need to differentiate between personal exposure of ambient- and non-ambient components (Rhomberg et al., 2011; Zeger et al., 2000). $PM_{2.5}$ is a complex mixture with chemical constituents, derived from various sources with different toxicities (Krall et al., 2017; Lakey et al., 2016; Pun et al., 2014; Stanek et al., 2011). People spend the majority (> 85%) of their daily time indoors in Hong Kong (Chau et al., 2002; Chen et al., 2018). The World Health Organization reported that indoor (household) air pollution has equal or more significant health risks compared with ambient air pollution (WHO, 2014). Past evidences suggests that ambient $PM_{2.5}$ infiltration rate vary across different chemical components (Chen et al., 2019; Ji et al., 2018). There were evidence that aside from particles of ambient origin, indoor sources (e.g., cooking, cleaning, exposure to tobacco smoke) and sources related to personal activity strongly affect total personal $PM_{2.5}$ exposures (Meng et al., 2009; Shang et al., 2019; Wan et al., 2011). Tailpipe exhaust, tire/brake wear, and road dust exhibited a spatially heterogeneous distribution (Krall et al., 2018). Personal exposure to traffic-related pollutants varied across individuals and (or) subpopulations (Baccarelli et al., 2014; Chen et al., 2018). These findings indicated that ambient concentrations at central monitoring stations may not adequately reflect the characteristics of personal exposures (Chen et al., 2017b; Kim et al., 2005; Koistinen et al., 2004; Larson et al., 2004; Shang et al., 2018; Shang et al., 2019).

Past studies indicated that human exposure to $PM_{2.5}$ component, including organic carbon (OC), elemental carbon (EC), sulfate (SO_4^{2-}), nitrate (NO_3^-), ammonium (NH_4^+) and elements (e.g., Na,

97 Si, K, Ca, Cu, Zn, V) was associated with increased cardiovascular and respiratory hospitalizations
98 and mortality (Achilleos et al., 2017; Pun et al., 2014; Tian et al., 2013). Source apportionment
99 techniques, such as Positive Matrix Factorization (PMF), Principal Component Analysis (PCA)
100 with Absolute Principal Component Scores (APCs) were applied on ambient and (or) indoor data
101 to estimate the contribution of specific sources to $PM_{2.5}$. Other studies conducted regression
102 analyses to differentiate the contribution of indoor and outdoor sources to particles in residential
103 houses (Hassanvand et al., 2014; Xu et al., 2014). A few studies also applied these methods to
104 determine the potential sources contributing to personal exposures (Hopke et al., 2003; Koistinen
105 et al., 2004; Larson et al., 2004; Minguillon et al., 2012). Most of these exposure studies were
106 conducted in North America and European cities (Johannesson et al., 2007; Weisel et al., 2004;
107 Williams et al., 2000). For example, Kim et al. (2005) applied the mixed-effects model to estimate
108 the relative contribution of primary sources of personal $PM_{2.5}$ exposures. Currently, a few studies
109 characterize the sources of personal $PM_{2.5}$ exposure for young students in Chinese megacities (e.g.,
110 Tianjin, Guangzhou) (Chen et al., 2017b; Zhang et al., 2015), with few studies fouced on the
111 seasonal or occupational differeces (Shang et al., 2019).

112 We conduct repeated personal monitoring in healthy adult subjects of Hong Kong. The results
113 regarding the determinants of personal exposure to $PM_{2.5}$ mass and its components were reported
114 elsewhere (Chen et al., 2018). Source contributions to personal $PM_{2.5}$ exposures were estimated
115 using PMF and PCA/APCs-linear mixed linear regression analysis. This study provides an
116 opportunity to leverage the advantage of different modeling techniques in source apportionment
117 of personal $PM_{2.5}$ exposure. Subsequently, seson and subject-group startified analysis regarding
118 source apportionment of personal $PM_{2.5}$ exposures were performed. The mixed source-specific
119 $PM_{2.5}$ contributes to elevated personal exposures, and the findings in this study provide

120 comprehensive information for epidemiologists and regulatory agencies regarding mitigation
121 strategies for particulate pollution.
122

123 ## 2. Materials and methods

124 2.1. Study subjects and protocol

125 We conducted repeated personal monitoring in 48 healthy adults (ages 18–63) in Hong Kong. All
126 subjects (including 12 office workers, 16 students, 12 housewives, and 8 non-office workers) were
127 non-smokers, not exposed to environmental tobacco smoke in indoor microenvironments (e.g.,
128 home, school, office, or other indoors), and did not have any chronic diseases. Personal samples
129 were collected between July 2014 and March 2015. Forty-two and 41 participants participated in
130 summer and winter sampling respectively; 35 of the 48 subjects participated in both sampling
131 seasons. A total of 161 personal samples were collected throughout the study period. The Joint
132 Chinese University of Hong Kong-New Territories East Cluster Clinical Research Ethics
133 Committee approved this study. Each subject signed informed consent before participatation.
134

135 2.2. Personal monitoring

136 Twenty-four hour (24-hr, 00:00–24:00 local time) personal $PM_{2.5}$ sample was collected using a
137 Personal Environmental Monitor (PEMs, Model 200, MSP Corp., Shoreview, MN, USA) together
138 with a Leland Legacy sampling pump (SKC Inc., Eighty-Four, PA, USA) operated at a flow rate
139 of 10 L/min. Two PEMs loaded with one Teflon membrane and one quartz fiber filter (37 mm, 2
140 μm pore size, Pall Corporation, MI, USA), respectively, were carried by each subject. Study
141 subjects were required to wear the PEMs near the breathing zone during each 24-hr sampling
142 session, except for sleeping and sitting, in which case the sampler was located in proximity to the
143 subjects (< 1 meter). Subjects were encouraged to maintain regular daily activity patterns.

144 Participants were also required to complete a 24-hr time-activity diary during each sampling
145 session (Tables S1-S2). A questionnaire regarding participants' personal information (i.e., age, sex,
146 occupation, and residential characteristics) was also acquired from each subject before
147 participating in this study. Fig. 1 shows the subjects' residential location in the study area in Hong
148 Kong. Further detail on quality assurance and quality control during personal monitoring and
149 sample collection is available elsewhere (Chen et al., 2017a).
150
151 2.3. Description of ambient data
152 Ambient $PM_{2.5}$ samples were acquired every sixth day at seven fixed monitoring stations in Hong
153 Kong (including roadside Air Quality Monitoring Site [AQMS] at Mong Kok; the urban AQMSs
154 at Central/Western [CW], Tsuen Wan [TW] and Kwai Chung [KC]; the new town AQMSs at Tung
155 Chung [TC] and Yuen Long [YL]; the suburban AQMS at Clear Water Bay [WB]) (Fig. 1). Daily
156 meteorological data including temperature (T), relative humidity (RH), rainfall (R), atmospheric
157 pressure (P), wind speed (WS), and wind direction (WD) retrieved from the Hong Kong
158 Observatory (HKO, http://www.weather.gov.hk/contente.htm) were assembled. The location of
159 the nearest weather stations for each AQMS is shown in Fig. 1.
160 Fig. S1 depicts the corresponding distances between seven central monitoring stations and
161 subjects' residences, with an average of ~14.9 km (range 0.1–34.5 km). In this investigation,
162 ambient data (269 sampling days, including $PM_{2.5}$ mass and constituents) for the overlapping
163 sampling period of July-October 2014 and December 2014-March 2015 were analyzed (Fig. S2).
164 Additional details about ambient data can be found in our recent publication (Chen et al., 2019).
165
166 2.4. Filter analyses

Triplicate filter weights (± 3 μg) were determined using an electronic microbalance (readability of 1 μg, Sartorius AG, Model ME 5-0CE, Goettingen, Germany) in a temperature (20–25 °C) and relativity humidity (35 ± 5%) controlled weighting room at The Hong Kong Polytechnic University, Hong Kong. Carbonaceous aerosols (organic carbon [OC] and elemental carbon [EC]) were analysed on quartz filters by the thermal/optical reflectance (TOR) method following the IMPROVE_A protocol using a DRI Model 2001 Thermal/Optical Carbon Analyzer (Atmoslytic Inc., Calabasas, CA, USA) (Chow et al., 2011). Method detection limits (MDLs) of OC and EC were 0.28 and 0.04 μg/m^3, respectively. Subsequently, water-soluble anions (including chloride [Cl^-], nitrate [NO_3^-], sulfate [SO_4^{2-}], and oxalate [$C_2O_4^{2-}$]) and cations (including sodium [Na^+], ammonium [NH_4^+], potassium [K^+], magnesium [Mg^{2+}], and calcium [Ca^{2+}]) were quantified using an ions Chromatograph (Dionex ICS-3000, USA) on these quartz filters (Ho et al., 2014). MDLs of ions were within the range of 0.01-0.23 μg/m^3.

Teflon filter samples were sent to Desert Research Institute laboratories (DRI, Reno, NV, USA) in a temperature-controlled cooler (< 4˚C). Elemental analysis by Energy Dispersive X-Ray Fluorescence (ED-XRF, Epsilon 5, PANalytical Company, Netherlands) was conducted (Watson et al., 1999). A total of 24 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], Rubidium [Rb], Strontium [Sr], Zirconium [Zr], Molybdenum [Mo], Tin [Sn], and lead [Pb]) were quantified (Chow and Watson, 2012). MDLs of analyzed elements were within the range of 0.5–33 ng/m^3 (Table S3). The analyzed 24 elements were detectable (i.e., > MDLs) for > 65% of the samples except Se, Mo and Sn (31-63% were detectable). Quality assurance (QA) and quality control (QC) protocols were followed when conducting personal monitoring (see Supporting

Information). Additional details about personal $PM_{2.5}$ gravimetric and chemical analyses can be found in our recent publication (Chen et al., 2018).

2.5. Source apportionment

We employed the U.S. Environmental Protection Agency's Positive Matrix Factorization (EPA PMF 5.0 software) receptor model to estimate sources contribution to personal $PM_{2.5}$ exposures in adult subjects in Hong Kong. Details about PMF modeling were described in (Hopke et al., 2003). $PM_{2.5}$ component concentrations, together with the uncertainties, were used as input in PMF analysis. Missing data were replaced by components median concentrations along with four times uncertainty; measured concentrations below MDLs were assigned as half of the MDLs value along with uncertainty of 5/6 MDLs (Reff et al., 2007). When there were redundant tracers, only one of them would be included. For example, Na and Ca were kept to prevent double counting in PMF analysis; for Cl, Mg, S, and K, the ions data (i.e., Cl^-, SO_4^{2-}, Mg^{2+}, K^+) were retained. We set 10% extra modeling uncertainty for PMF analysis. We applied error estimation (EE) diagnostics to determine the optimal number of source factors. In the present study, seven source factors (F_{peak} = -0.5) were selected in PMF analysis, which was optimal and balancing the statistical robustness and physical interpretability. Detailed information related to EE diagnostics can be found in Brown et al. (2015).

PCA was applied to identify major sources contributing to $PM_{2.5}$. A Varimax normalized rotation was performed to maximize or minimize the factor loadings of principal components (PCs). The principal component with an eigenvalue greater than one entered in the source identification analysis. The absolute principal component scores (APCs) for each component was calculated by subtracting the absolute zero factor sources from the corresponding factor scores in the standard

PCA analysis (Guo et al., 2004; Thurston and Spengler, 1985). We applied PCA/APCs incorporated with linear mixed-effects models (LMM) to quantify sources contribution of personal exposure to $PM_{2.5}$. The PCA/APCs-linear mixed effects model (LMM) expressed as follows:

$$Y_{ij} = \beta_0 + \sum_{m=1}^{k} \beta_m APCs_m + b_i + \varepsilon_{ij} \quad \text{Eq. 1}$$

where Y_{ij} is the exposure level on the *j*th day of *i*th subject, β_0 is the intercept of the regression for the exposures, β_m represents the regression coefficient for the $APCs_m$; b_i represents the random effect for *i*th subject, and ε_{ij} refers to the random effects of exposures on *j*th day for *i*th subject. In model (1), the random effects of b_i and ε_{ij} are presumed to mutually independent with means of zero and between-subject (σ^2_b) and within-subject (σ^2_w) variance. Subsequently, the mixed-effects regression analyses were stratified by season to account for the heterogeneity of the source contribution of personal $PM_{2.5}$ exposure using backward stepwise regression method to eliminate non-significant ($p > 0.05$) variables. Also, we divided all subjects into two groups, group A (combining categories: office worker and student) and group B (housewife and non-office worker), instead of four occupations to account for heterogeneity of source contributions between groups.

2.6. Statistical analysis

Statistical differences in personal exposure to $PM_{2.5}$ components were evaluated using analysis of variance (ANOVA) by season and by subject group. We utilized the same dataset to conduct both PMF and PCA/APCs-LMM (linear mixed-effects model) analysis and to compare modeling results. Pearson's correlation (*r*) was applied to characterize the magnitude of associations between chemical constituents in personal $PM_{2.5}$. A *p-v*alue < 0.05 was considered as statistically significant. We illustrated linear mixed-effects regression models using *lemr* from the *lme4* and *lmerTest* package in R 3.4.4 (The R Project for Statistical Computing, 2018: http://www.r-

project.org) (Bates et al., 2014). We used the marginal R^2 statistic (R^2_m) to estimate the overall predictive ability of the mixed-effects model; semi-partial R^2 were calculated for each APCs in the mixed-effects model (Jaeger, 2016; Jaeger et al., 2016).

3. Results

3.1. Characteristic of personal exposure to $PM_{2.5}$ components

The mean and standard deviations for the analyzed $PM_{2.5}$ components in personal samples are shown in Tables 1 and 2 by season and by subject group. Attributed to the extra trace elements included (e.g., Se, Rb, Sr, Zr, Mo, Sn) in this analysis, there were slight mass differences between the means of the elements reported in this study and those listed in our previous publication in (Chen et al., 2018).

OC and EC accounted for 24.3 ± 10.3% and 7.2 ± 5.7% of personal $PM_{2.5}$ mass, respectively, with no seasonal differences ($p > 0.05$). SO_4^{2-} was the most abundant component, accounting for 26.4-28.9% of personal $PM_{2.5}$ mass, followed by NH_4^+ and NO_3^- representing about 7.2-24.3% and 7.0-11.1% of the personal $PM_{2.5}$ mass, respectively. All water-soluble ions exhibited significant seasonal variations ($p < 0.01$) with higher concentrations in winter and lower levels in summer except Na^+ ($p = 0.61$). Personal Ca^{2+} exposure was higher in summer than in winter ($p = 0.31$). Total elements (7.0–8.7 ng/m^3) accounted for 22.1 ± 6.8% of personal $PM_{2.5}$ mass and exhibited significant seasonal variations ($p = 0.02$). The reconstructed mass was strongly correlated with the measured mass for personal $PM_{2.5}$ ($r > 0.95$; $p < 0.01$) (Chen et al., 2018).

Higher personal OC and EC exposures were observed in housewives ($p = 0.004$) and non-office workers ($p < 0.001$) compared to other subjects. Also, water-soluble ions and most of the trace elements (e.g., Al, S, K, As, Rb, Sr) concentrations were higher in housewives and non-office

workers in winter than in their counterparts. Fig. 2 illustrates the personal$_{_avg}$ to ambient$_{_avg}$ ratios for $PM_{2.5}$ components across seasons. Fig. S2 shows the $PM_{2.5}$ component concentrations in ambient samples during the same study period. OC, Na^+, secondary inorganic ions（SIA, including SO_4^{2-}, NO_3^-, NH_4^+）, Ca, Ti, Fe, Cu, Rb, Zr, and Sn exhibited higher levels in personal samples than those of ambient, with personal$_{_avg}$/ambient$_{_avg}$ ratios greater than unity in both seasons.

3.2. Source apportionment of personal $PM_{2.5}$ exposure using PMF

Fig. 3 shows the estimated $PM_{2.5}$ source profiles of personal exposure, depicted as percentage of variables to each factor derived from the PMF model. The results were robust with an R^2 value of 0.9 ($p < 0.001$) between experimental concentrations and the fitted values. Factor 1 had high loadings for V and Ni; these two elements were the dominant species from oil combustion and were considered as robust tracers from shipping emissions in coastal areas (Oeder et al., 2015; Tao et al., 2017). Factor 2 had high loadings for secondary nitrate, chloride, and magnesium. Factor 3 characterized by Al, Si, Ca, Ti, and Sr, that we named crustal/road dust (Cesari et al., 2016; Ho et al., 2003b). Factor 4 was identified as non-tail pipe source derived from traffic, characterized by high loadings of Fe, Cr, and Mn (Moreno et al., 2018). Factor 5 was characterized by high loadings for sulfate, ammonium, and oxalate. Factor 6 was interpreted as tailpipe exhaust, characterized by high loadings of OC and EC (Schauer, 2003). Factor 7 was identified as a composite of two sources from regional pollution that could not be further separated, including biomass burning (based on the abundance of K^+) (Sang et al., 2011) and coal combustion from power plants and industrial facilities (based on the abundance of Cu, Zn, As, Br, Pb) (Pun et al., 2015). Fig. 4 illustrates the contributions of the seven identified sources to personal $PM_{2.5}$ exposures. Secondary sulfate accounted for the largest fraction (25.9%) of total personal $PM_{2.5}$ exposures, followed by regional

pollution (21.9%), tailpipe exhaust (19.0%), secondary nitrate + Cl^- (15.1%), shipping emissions (8.1%), crustal/road dust (5.2%) and non-tailpipe particles (4.7%).

Fig. S3 illustrates the sources profiles extracted from the ambient samples. Personal and ambient data showed similar sources profiles, with differences being observed for traffic-related particles in personal samples. Further, factor 8 had high loadings of Na (> 80%) and Cl^- in ambient $PM_{2.5}$ (Fig. S3), which was not shown in personal samples. The contribution of each source to ambient $PM_{2.5}$ followed the same order as those for personal $PM_{2.5}$ exposures, except marine aerosol and shipping emission that accounted 15.6% and 1.1% of ambient $PM_{2.5}$, respectively (Fig. S4).

3.3. Source apportionment of personal $PM_{2.5}$ exposure using PCA/APCs–LMM

Fig. 5 shows the correlation matrix for personal $PM_{2.5}$ components. Table 3 lists the rotated component matrix (i.e., factor loadings) from PCA in personal $PM_{2.5}$. Five PCs explained 79.1% of the total variance in this study (Table 3). PC1 (explained 28.2% of the original data variance) was characterized by high factor loadings of Al, Si, Ca, and Ti and to a lesser extent, Mn, Fe, Zn, Rb, Sr, and Zr (Table 3). Further, PC1 and PC3 (characterized by high loadings of V and Ni) (Table 3) closely resembled the corresponding PMF sources profile for Factor 3 and Factor 1 (Fig. 3), respectively. PC2 explained 28.1% of the total variance and illustrated mixed profiles with secondary nitrate (Factor 2), secondary sulfate (Factor 5), regional pollution (Factor 7), and tailpipe emission associated species (i.e., EC). PC4 included Cl^- and a lower loading of secondary nitrate. PC5 characterized by Cr and Fe (accounted for 5.8% of the total variance) and interpreted as non-tailpipe pollutants.

Table 4 displays the source contributions of personal $PM_{2.5}$ exposures by using PCA/APCs-LMM model. The modeled personal $PM_{2.5}$ exposures were highly correlated with the measured

values ($R^2 = 0.87$; $p < 0.001$). On average, a combination of regional pollution, secondary nitrate and sulfate, and tailpipe exhaust contribute to 61.7% of personal $PM_{2.5}$ exposure. Crustal/road dust and shipping emissions are accounting for 9.3% of personal $PM_{2.5}$; lesser contributions were shown for traffic-related particles (4.3%). PC4 (secondary nitrate + Cl^-) showed considerably lower contribution (3.7%) to personal $PM_{2.5}$ because it mixed with other sources. Component matrix from PCA in ambient $PM_{2.5}$ and sources contributions of ambient $PM_{2.5}$ using PCA/APCs-LMM model are shown in Tables S4–S5.

3.4. Seasonal variation and subject-specific source contributions to personal $PM_{2.5}$ exposure

Source contributions to personal $PM_{2.5}$ exposures were stratified by season and by study group. Six PCs (with eigenvalues greater than 1) were extracted by season (Tables S6–S7) while 5 PCs were identified across different groups of subjects (Tables S8–S9). Although the order of PCs variated, the results shared a similar number of source factors and the total variance explained by the PCs was comparable across seasons (79.5–85.7%) and subject groups (75.8-83.4%).

Table 5 and Fig. 6 show the estimated seasonal- and group-specific source contributions to personal $PM_{2.5}$ exposure using PCA/APCs–LMM analysis. Tailpipe exhaust (Factor 6) and traffic-related particles (Factor 4) contributing to 52.6% and 6.7% of personal $PM_{2.5}$ exposure in summer, which were both significantly higher ($p < 0.001$) than those in winter (< 37.4% and 3.0%, respectively). In contrast, the contribution of shipping emission to personal $PM_{2.5}$ exposure was significantly higher in winter than in summer (8.3% vs. 7.0%; $p < 0.001$). The least variability was found for the contributions of the combination of secondary aerosols and regional pollution (Fig. 6). Shipping emissions are contributing to 6.2–8.4% of total personal $PM_{2.5}$ across all subjects.

Two opposite trends were shown: group A with secondary aerosol (39.6%) contribution maximum and tailpipe exhaust (40.2%) exhibited an increase in group B (e.g., van-drivers, paper vendors).

4. Discussion

In this study, we utilized the personal exposure data collected in adult subjects in Hong Kong to estimate source contributions of personal $PM_{2.5}$ exposure. Similar seasonal variations were shown for both personal $PM_{2.5}$ mass and most of the personal $PM_{2.5}$ components, with higher levels in winter and lower levels in summer. For example, significantly higher personal SIA exposures were found in winter than those in summer ($p = 0.001$). In this analysis, the mean NO_3^-/SO_4^{2-} ratios in personal samples ranging from 0.31 to 0.43 by season, indicating the predominance of stationary sources (e.g., emission from sulfur-containing coal combustion) over mobile sources in personal $PM_{2.5}$ exposure in Hong Kong. The results were comparable to those in Guangzhou, which reported NO_3^-/SO_4^{2-} ratios of 0.3–0.4 in personal $PM_{2.5}$ exposure (Chen et al., 2017b). Strong correlation coefficients were shown between ammonium with sulfate ($r = 0.93$; $p < 0.01$) and oxalate ($r = 0.81$; $p < 0.01$). Coal-combustion related particles (i.e., S, As, Se, Pb) were significantly higher in winter than those in summer for personal $PM_{2.5}$ exposures. We also found that sulfate was highly correlated with As and Pb (r: 0.59–0.68; $p < 0.01$), indicating a coal combustion signature (Fig. 5). Higher personal SIA exposures along with $personal_{\_avg}/ambient_{\_avg}$ ratios > 1 for SIA may be attributed to outdoor particles infiltration and accumulation of indoor-generated particles. Moderate to strong correlations (R^2: 0.56–0.63; $p < 0.01$) between time in residence and personal SIA exposure were shown (Chen et al., 2018). The coefficient of divergence (COD) value for the average sulfur in ambient $PM_{2.5}$ was 0.16 (e.g., < 0.20), indicating the uniform distribution of sulfur over the study area in Hong Kong (Fig. S5).

Weak correlations were observed between EC and other analyzed components (except for OC, $r = 0.67$; $p < 0.01$). OC was also strongly associated with K^+, tracer for $PM_{2.5}$ from biomass burning emission ($r = 0.61$; $p < 0.01$) (Fig. 5). Comparable OC and EC mass fractions in personal $PM_{2.5}$ were shown across seasons. In contrast, average personal OC exposure decreased by 25.0-36.1% in office workers compared with other subjects. Significant differences in personal EC exposures were shown across different subject groups, with an exposure range of 1.7 ± 0.9 to 3.0 ± 1.2 μg/m^3 ($p < 0.001$). Past studies pointed out that personal EC exposure associated with individuals' activity patterns (Huang et al., 2015; Tunno et al., 2016). Subject-specific activity patterns are summarized in Tables S8–S9 by season. In our recent publication, we found that for one-hour extra time in transit an average increase of 0.47 μg/m^3 (95% CI: 0.36–0.67 μg/m^3) in personal EC exposure was shown (Chen et al., 2018). Dons et al. (2011) indicated that the transport microenvironment was one of the most important contributors to personal EC exposure, for instance, 6% of the time in transport contributed over 20% of total exposure.

The average Fe/Ca (1.89) and Mn/Ca (0.08) ratios in personal $PM_{2.5}$ were comparable with the corresponding values in soil and paved road dust samples (0.8-2.6 and 0.01-0.08, respectively) in Hong Kong (Ho et al., 2003a; Ho et al., 2003b). Personal exposure to OC, Ca, Ti, Fe, Se, Rb, Sr, Zr were significantly higher than those in ambient $PM_{2.5}$, with personal$_{_avg}$/ambient$_{_avg}$ ratios greater than unity in both seasons (Fig. 2). Further, personal exposure to crustal/dust particles (Ca^{2+}, Mg, Si, Ca, Ti) and non-tailpipe traffic-related particles (Mn, Fe, and Zn) did not exhibit apparent seasonal trends. These sources contributed to personal $PM_{2.5}$ due to exposure when subjects stayed indoors, near roadways, and in transit (e.g., bus, metro). This agreed with previous findings (Dons et al., 2011; Spira-Cohen et al., 2010), which suggested that indoor exposure to ambient elements (e.g., Mn, Fe, Sr, Mo) significantly decrease with road proximity (Huang et al.,

2018). Also, the previous finding showed that increment (per hour) of time in transit associated with increased personal exposure to Ti and Fe ($p < 0.05$) in adults in Hong Kong (Chen et al., 2018).

In this investigation, strong and statistically robust correlations were shown (r: 0.56-0.91; $p < 0.01$) between crustal/road dust and non-tailpipe pollutants (Fig. 5). Personal exposure to crustal/road dust particles and non-tailpipe pollutants varied across subject groups. Personal EC, Ca^{2+}, and Sr exposures were significantly higher ($p < 0.05$) for non-office workers than other subjects. These findings are consistent with previous studies. For example, Huang et al. (2014) reported significantly higher crustal particles (Si, Al, Ca, Ti) among truck drivers than those in office workers in Beijing, and provided evidence showing that these crustal particles associated with decreased lung functions in truck drivers. It should be noted that the highest ambient levels (43.2 μg/m^3, $p = 0.02$) coincided with the significantly higher $PM_{2.5}$ components exposures ($p < 0.05$) for housewives compared with other subjects (Table 2). No clear correlation was observed between the analyzed $PM_{2.5}$ components exposure levels with any of our recorded indoor activities.

Agrawal et al. (2009) reported V/Ni ratio > 2.2 in heavy fuel oil. Wang et al. (2018) showed average V/Ni ratio of 3.1 in the Port of Shanghai. In this study, V/Ni ratio in ambient $PM_{2.5}$ varying from 2.9-3.3 with an average of 3.2 across the seven ambient sites. A relatively higher V/Ni ratio of 4.5 was shown in personal $PM_{2.5}$ throughout the sampling period. Similar results were obtained in ambient $PM_{2.5}$ in Spain with V/Ni ratio of 4 ± 1 (Viana et al., 2009). The results above agreed with the previous findings (Viana et al., 2014). A strong correlation coefficient ($r = 0.96$; $p < 0.01$) between Ni and V was shown with personal$_{_avg}$/ambient$_{_avg}$ ratios < 1 for both species, indicating the predominance of the ambient source from shipping emissions.

PMF identified six sources including regional pollution, secondary sulfate, secondary nitrate, tailpipe exhaust, crustal/road dust, and shipping emission for both ambient and personal datasets. The sources represented by regional pollution, secondary sulfate, and secondary nitrate contributed 55.7% of ambient $PM_{2.5}$, which was considerably lower than that in personal $PM_{2.5}$ exposures (62.9%). In a previous study conducted by (Huang et al., 2013), whose findings consistent with the current study, they revealed that secondary sulfate (31%), regional pollution (mainly biomass burning) (23%), secondary nitrate (13%) were three dominant contributing sources to ambient $PM_{2.5}$ at the suburban AQMS in Hong Kong. Lu and Fung (2016) pointed out that superregional transport (e.g., power plant, industry emissions) over Hong Kong was an important contributor to sulfates (73–92%) and nitrates (30–76%) in both summer and winter season. In our recent publication, we found that time spent indoors at home associated with personal exposure to SIA (Chen et al., 2018). Significant higher personal SIA exposures were found in housewives compared with other subjects because they spent more time indoors (86.0-90.5%) than their counterparts (47.9–74.2%).

The sources represented by tailpipe exhaust, crustal/road dust contributed 18.2% and 9.4% of ambient $PM_{2.5}$, which were also lower than that for personal $PM_{2.5}$ exposure (19.0% and 9.9%, respectively). For personal $PM_{2.5}$ exposure, the dust-related pollution was separated into two parts (i.e., 5.2 % of crustal/road dust, 4.7% of the traffic-related fraction), reflecting the influence of personal activity (e.g., in transit) on total exposure levels. The latter reflected personal exposure to particles of non-ambient origin (e.g., indoor-generated particles, personal activities while indoors and outdoors).

Although there were discrepancies in the number of factors (5 or 6 vs. 7), the most identifiable PMF factors agreed well with the identified PCs. Furthermore, most of the PMF factors were easily

418 distinguishable from others by notable differences in the source profiles. For example, concerning
419 the PCA, the PMF model can distinguish between the traffic-related particles and crustal/road dust,
420 and separate secondary sulfate, secondary nitrate, and regional pollution. Our results consistent
421 with the findings in (Cesari et al., 2016; Ito et al., 2004), that conducted inter-comparison of source
422 apportionment in ambient $PM_{2.5}$ and PM_{10} using both PCA and PMF model.
423 PCA/APCs–LMM analysis revealed that regional pollutants + secondary aerosol + tailpipe
424 exhaust (65.4%), shipping emission (9.3%), traffic-related particles (9.3%) and crustal/road dust
425 (4.3%) were positive and significant contributors of personal $PM_{2.5}$ exposure. This agreed with
426 previous findings in Guangzhou (Chen et al., 2017b), which showed that regional air pollution
427 (50.4 ± 0.9%), traffic-related particles (8.6 ± 0.7%), dust-related particles (5.8 ± 0.7%),
428 and biomass burning emissions (2.0 ± 0.2%) were positive sources of personal $PM_{2.5}$ exposure in
429 Guangzhou.
430 Subsequently, we applied PCA/APCs–LMM analysis to characterize the seasonal and
431 occupational variations of sources contributions to personal $PM_{2.5}$ exposure. The mixed-effects
432 model was constructed with personal $PM_{2.5}$ exposure as the dependent variable and APCs as the
433 independent variables. The APCs were regressed against personal $PM_{2.5}$ exposure to estimate
434 source contributions. The contribution of each source was calculated by multiplying the regression
435 coefficients from the mixed-effects model by their respective concentrations and dividing by the
436 fitted values. Backward elimination resulted in a multi-sources regression model (Tables 4-5). The
437 results showed that fresh sea-salt (Cl^-) was not a significant contributor to personal $PM_{2.5}$ exposure
438 compared with other sources in summer. A portion of regional pollution (mainly industrial
439 activities, with high loadings of Mn and Pb) contributes to 5.5% of personal $PM_{2.5}$ in winter.
440 Moreover, secondary aerosols mixed with regional pollutant contributed 34.6-43.6% of personal

441 $PM_{2.5}$ exposure with no seasonal or occupational differences ($p > 0.05$). Previous studies suggested
442 that variations of regional pollutants were mostly due to the influence of temporal variability in
443 the ambient environment (Minguillon et al., 2012).

444 In this investigation, the source contribution of tailpipe exhaust to personal $PM_{2.5}$ exposure was
445 exceptionally high, reaching 50% in summer ($p < 0.001$). Huang et al. (2013) indicated that vehicle
446 emission was more dominant during sampling periods with lower $PM_{2.5}$ concentrations in Hong
447 Kong. In addition, the contribution of tailpipe exhaust to personal $PM_{2.5}$ exposure varied widely
448 across subject groups (< 33.3% vs. 40.5%; $p < 0.05$). This may be attributed to the variation of
449 time in transit (Chen et al., 2018). For example, non-office workers spent about 4.2–4.6 hour (i.e.,
450 17.3–19.3%) of daily time in vehicle, which significantly higher than their counterparts (range of
451 1.4–4.4%, $p < 0.001$) (Table S8-S9). Expected higher non-tailpipe traffic contribution (8.2%) to
452 personal $PM_{2.5}$ exposure was also found in group B (mainly in non-office workers) compared to
453 group A. For all subjects, the amount of time in transit was higher in summer (5.9 ± 10.0%) than
454 that in winter (4.0 ± 7.8%). Traffic-related particles contributing 6.7% of personal $PM_{2.5}$ exposure
455 in summer, which was significantly higher ($p < 0.001$) than that in winter (3.0%).

456 Previous studies targeted the contribution of shipping emissions to ambient pollution without
457 considering individual or population exposure. For example, Pun et al. (2014) pointed out that
458 residual oil combustion (characterized by Ni and V) contributed 8.0% of ambient $PM_{2.5}$ in Hong
459 Kong. Tian et al. (2013) linked elevated ambient Ni and V concentrations with increased
460 cardiovascular hospitalizations in Hong Kong. Lin et al. (2018) indicated that each IQR increase
461 (median = 1.0 $\mu g/m^3$) in lag_1 shipping emission was associated with a 5.6% (95% CI: 0.8-10.5%)
462 increment in cardiovascular mortality in Guangzhou, China. This investigation is the first study

463 that used repeated personal measurements to quantify the shipping emission contribution to
464 personal $PM_{2.5}$ exposure.

465 Concurrent ambient $PM_{2.5}$ species were not available in this investigation. Thus ambient data
466 during the same sampling period were used for comparison. In this analysis, we reported the
467 seasonal $personal_{\_avg}/ambient_{\_avg}$ ratio instead of daily personal/ambient ratios for $PM_{2.5}$
468 components. There were some limitations: 1) although repeated personal monitoring was
469 conducted, exposure error cannot be eliminated as a possibility; 2) analytical uncertainties for each
470 species might also influence the results; 3) the small number of subjects may limit the prediction
471 power, and it may also limit the generalizability to other cities or subpopulations. Even with these
472 limitations, the model structure and the use of source profiles constraints provide a useful tool for
473 evaluating various sources contribution to personal $PM_{2.5}$ exposure. These two approaches (PMF
474 vs. PCA/APCs-LMM) are complementary and allowed for the evaluation of dataset collectively
475 and separately. Further investigation of the complex relationships between long-term source-
476 specific pollution exposure and health effects are needed for better air pollution control policies
477 and evidence-based public health interventions.

478

479 ## 5. Conclusions

480 Determining source contributions of individual or population exposure remains a challenge in
481 epidemiological studies. We applied source apportionment technique including PMF and
482 PCA/APCs-LMM analysis to investigate and to explain variability in sources contributions to
483 personal $PM_{2.5}$ exposure in adult subjects in Hong Kong. We identified seven source profiles for
484 personal $PM_{2.5}$ exposure using PMF. This study revealed that the combination of PCA/APCs with
485 LMM analysis could be used to determine source contributions to personal $PM_{2.5}$ exposure, as well

as to assess the seasonal and occupational variations in source contributions. We found that regional pollution and tailpipe exhaust contributed to $50.4 \pm 0.9\%$ and $8.6 \pm 0.7\%$ of personal $PM_{2.5}$ exposure. Our findings also highlighted the significance of traffic-related particles (4.7%) and shipping emissions (8.1%) to personal $PM_{2.5}$ exposures. The most crucial difference between personal and ambient modeling results was that traffic-related particles lead to a positive and significant role in influencing personal $PM_{2.5}$ exposure. Different source apportionment approaches for investigating sources contribute to total personal exposure can be further applied to source-specific health risks assessment.

Author Contributions

XCC was involved in data analysis and manuscript preparation. TW and KFH designed the research. XCC, JJC, and SCL performed the sample collection and laboratory work. TW, HLY, and KFH revised the manuscript. HLY and NCL supervised the development of study and manuscript evaluation. All authors read and approved the final manuscript.

Conflict of interest

The authors declare that they have no competing interests.

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