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Personal Exposure of Particulate Matter in Hong Kong

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1. INTRODUCTION

1.1   Background

Air pollution is an ongoing issue and poses a significant threat to human health worldwide. Fine particles ($PM_{2.5}$, particles with an aerodynamic diameter less than or equal to 2.5 µm) is an important air pollutant, which is proven to be associated with different adverse human health effects such as cardiovascular and respiratory morbidity and mortality (WHO, 2013; Kim et al., 2015). Attention in the scientific community has been focusing on discrepancies between short-term (hours, days) exposure effects and long-term (months, years) cohort studies (Beverland et al., 2012). Numerous epidemiological studies reported that there are significant associations between short-term and long-term exposure to $PM_{2.5}$ and are the main reasons for causing adverse human health effects (Adren et al., 2004; Analitis et al., 2006). The World Health Organization (WHO) has estimated that approximately 3.7 million deaths and 4.3 million deaths globally in 2012 could be attributed to ambient air pollution and indoor air pollution, respectively (WHO, 2012; WHO, 2014). Epidemiological studies have already shown associations between $PM_{2.5}$ concentrations and population mortality also at levels well below the WHO guideline level for $PM_{2.5}$, without evidence of a threshold of exposure to $PM_{2.5}$ below which no adverse health effects would be anticipated.

Previous epidemiological studies are usually based on outdoor air concentration data collected at fixed ambient air quality monitoring sites, however, these concentrations alone are not sufficient for a thorough understanding of the cause of adverse human health effects due to people spend 80-90% of their time in the indoor environment (Cao et al., 2005). Previous studies demonstrated that outdoor air pollutant concentrations vary from indoor leading to different human health outcomes (Ji and Zhao, 2015). This difference is a result of different cumulative indoor sources and personal exposure activities (Wilson and Brauer, 2006). Results in the past, in general, showed positive correlations between $PM_{2.5}$ exposure and adverse health effects with the correlations vary from geographic locations due to different PM composition (Chung et al., 2015). In modern day, many atmospheric chemistry studies have been put emphasis on characterizing the PM composition in addition to investigating underlying toxicological

mechanisms using various biological exposure experiments. These experiments showed components such as organic carbon (OC), elemental carbon (EC), ions and heavy metals are linked to adverse human health outcomes (Rohr and Wyzga, 2012). Over the past three decades, the Pearl River Delta (PRD) region has been on a rapid pace of economic development and PM pollution is a prime environmental problem in the region. The PM sources in Hong Kong have long been known to be associated with motor vehicles emission, urban construction, cooking and trans-boundary pollution from the PRD region (Louie et al., 2005; Ho et al., 2006a). There is currently a lack of personal $PM_{2.5}$ exposure data in this city and the PRD region (Jahn et al., 2013). With the huge challenge of ensuring air quality in Hong Kong, it is essential to conduct an advanced personal exposure sampling campaign, acquiring information about OC/EC, water-soluble ions, and element content in the $PM_{2.5}$. This information, coupled with epidemiological studies will outline a bigger picture for the government and policymaker setting up suitable regulatory strategies safeguarding population health.

1.2   Long-term Impact

Its relevance, significance and value:

1. There is currently a lack of personal $PM_{2.5}$ exposure levels and their chemical composition information in Hong Kong. Data obtained from this study will identify $PM_{2.5}$ and their chemical components (e.g., OC/EC, water-soluble ions, metal elements) in the context of unique personal exposure to $PM_{2.5}$ via direct measurement.

2. Since the effects of personal exposure to $PM_{2.5}$ continue to receive much attention within different local scientific communities and regulatory bodies, this study will address several critical issues about their relationship between personal exposure in an urban environment and outdoor $PM_{2.5}$ concentrations in Hong Kong.

3. The outcome of this project will be used for further risk assessment and health related studies provided the information will be an integral part of effective public health protection strategy development in future.

1.3   Study Objectives

The objectives of this study are to characterize the seasonal and spatial variation of personal exposure to $PM_{2.5}$ and their corresponding chemical composition in Hong Kong. The relationship between personal $PM_{2.5}$ exposures and ambient $PM_{2.5}$ concentrations will also be assessed.

1.4   Scope of Study

The scope of this study was:

1. To collect $PM_{2.5}$ personal exposure samples from recruiting subjects inhabiting in different areas of Hong Kong and perform personal $PM_{2.5}$ gravimetric and chemical analyses;

2. To conduct questionnaire surveys and assess exposure characters of individual subjects; and

3. To assess the relationship between personal $PM_{2.5}$ exposures and ambient $PM_{2.5}$ concentrations and access their contribution to personal exposures.

This final report outlines a technical approach for investigating the personal exposure to $PM_{2.5}$ in Hong Kong. The report further characterizes the seasonal and spatial variations of personal exposure to fine particles ($PM_{2.5}$) and their chemical composition (e.g., OC/EC, water-soluble ions, elements) in Hong Kong. A one-year long (i.e., spring, summer, autumn, winter) personal $PM_{2.5}$ exposure monitoring campaign was illustrated in this report.

2. RESEARCH PLAN AND METHODOLOGY

2.1 Program Plan

- No less than fifty-two (52) non-smoking subjects of different gender, age, occupation and residential areas were recruited.

- To investigate seasonal and spatial variation, in addition to working/non-working day effect (e.g., Monday-Friday, Saturday-Sunday) of personal exposure to $PM_{2.5}$, 40 subjects were recruited from different areas in Hong Kong and conducted with twenty-four hours (24-hr) personal exposure monitoring including one day between Monday and Friday and another sampling day either on Saturday or Sunday during summer and winter, respectively.

- To investigate the association between personal exposures and ambient $PM_{2.5}$ concentrations, 12 subjects were recruited for personal exposure to $PM_{2.5}$ monitoring in one-year duration, and they were divided into three sampling groups (e.g., each group consists of four subjects). The sampling period was once in the every-18th-day cycle for each group in one year. The sampling date is in accordance with the ambient $PM_{2.5}$ sampling schedule of Hong Kong Environmental Protection Department (HKEPD). Personal exposure to $PM_{2.5}$ was monitored by subjects along with seven monitoring stations being established at different areas in Hong Kong and maintained by HKEPD (Section 2.2).

- The collected personal exposure samples were transferred to laboratories for gravimetric, organic carbon/elemental carbon, water-soluble inorganic ions, and elemental analysis.

- Questionnaires contained subject's personal information, and time-activity diaries were collected and assessed with personal exposure measurements by different correlation and regression methods.

2.2 Technical Approach and Methodology

In response to the Contract (Ref. 13-04909) requirement, the research team has duly obliged by providing the necessary expertise, materials, and equipment to accomplish the personal exposure measurement within the time and budget limit.

During the personal sampling period from April 2014 to June 2015, 24-hr ambient $PM_{2.5}$ mass measurements were acquired once every six days from the roadside station Mong Kok (MK), the urban Central/Western (CW) and Tsuen Wan (TW) Air Quality Monitoring Site (AQMS), the new town Tung Chung (TC) and Yuen Long (YL) AQMS, general station Kwai Chung (KC), and the Suburban Clear Water Bay (WB) Air Quality Research Site (AQRS) (Website: http://www.aqhi.gov.hk/en.html). Both Teflon-membrane and Quartz-fiber 47 mm filters were used at each sampling site to obtain ambient $PM_{2.5}$ samples. All collected ambient samples were analyzed for mass by gravimetry at HKEPD and subjected to chemical analysis, including 1) organic and elemental carbon (OC/EC) analysis; 2) ionic measurements and 3) measurements of elements from Sodium (Na) to Uranium (U).

2.2.1 *Recruitment of Subjects*

No less than fifty-two non-smoking subjects with different gender (i.e. the gender ratio in approximately 1:1 distribution), age, occupation, and residence areas were recruited based on non-probability sampling method. Potential subjects were excluded if any of these following criteria persisting: (1) Smoker; (2) Under 18 years of age; (3) Not residing in Hong Kong over the past twelve months; (4) Not in optimum physical condition (Diagnosis with disease such as chronic respiratory disease, coronary artery disease and cancer), and/or unable to carry the PEM. Occupation of all participants was categorized into different groups, e.g., office workers, non-office workers, housewives and students.

2.2.2 *Research Ethics*

Research ethics approval letter for the survey and behavioral research was granted by the Survey and Behavioral Research Ethics Committee at The Chinese University of Hong Kong (CUHK) for observation of human behavior in experiment prior personal exposure sampling period (Appendix A). This letter was to ensure that the ethical concern of participatory rights in the research was fully considered and the research was conducted in compliance with local legal codes (e.g., personal data, privacy ordinance).

The research team has completed human behavior research in the past with a

proven track record of handling privacy of research individual. Each participant was briefed about the proposed study and was required to sign a consent form in Appendix B. Chinese version of the consent form was given to the non-native English speaking participants. The consent form was completed before the beginning of the sampling process.

2.2.3 *Questionnaire*

All participants were interviewed and asked to complete a detailed questionnaire at home regarding with their corresponding medical conditions. Information about socioeconomic status and lifestyle was also collected. Factors associated with air pollutant exposure, such as transport time between office and home, environmental tobacco smoke (ETS), residence duration, a period of home renovation, cooking/heating and use of incense/mosquito coils were recorded. A sample questionnaire is attached in Appendix C.

2.2.4 *Personal PM$_{2.5}$ Monitoring*

Personal exposure sampling campaign was arranged as follows:

Schedule A

Twenty-four hours personal PM$_{2.5}$ samples were collected by each of the subjects in one weekday (i.e. Monday to Friday) and one day during the weekend (i.e. Saturday to Sunday) from 8th of July to 24th of October 2014 and from 9th of December 2014 to 37th of March 2015. A total of 90 and 82 sets of filter samples was collected during the first (defined as summer) and second (defined as winter) sampling campaign, respectively. Each set of PM$_{2.5}$ personal exposure samples consisted of a Teflon and one quartz filter sample. Field blanks were collected without switching on 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 campaign, respectively. Two subjects (EP04 and EP13) collected four sets of personal exposure samples while another two subjects (EP05 and EP21) collected three sets of samples during the summer campaign to account for the within-subject factor over time. The participants' resident locations for the first and second campaign are shown in Figure 2-1.
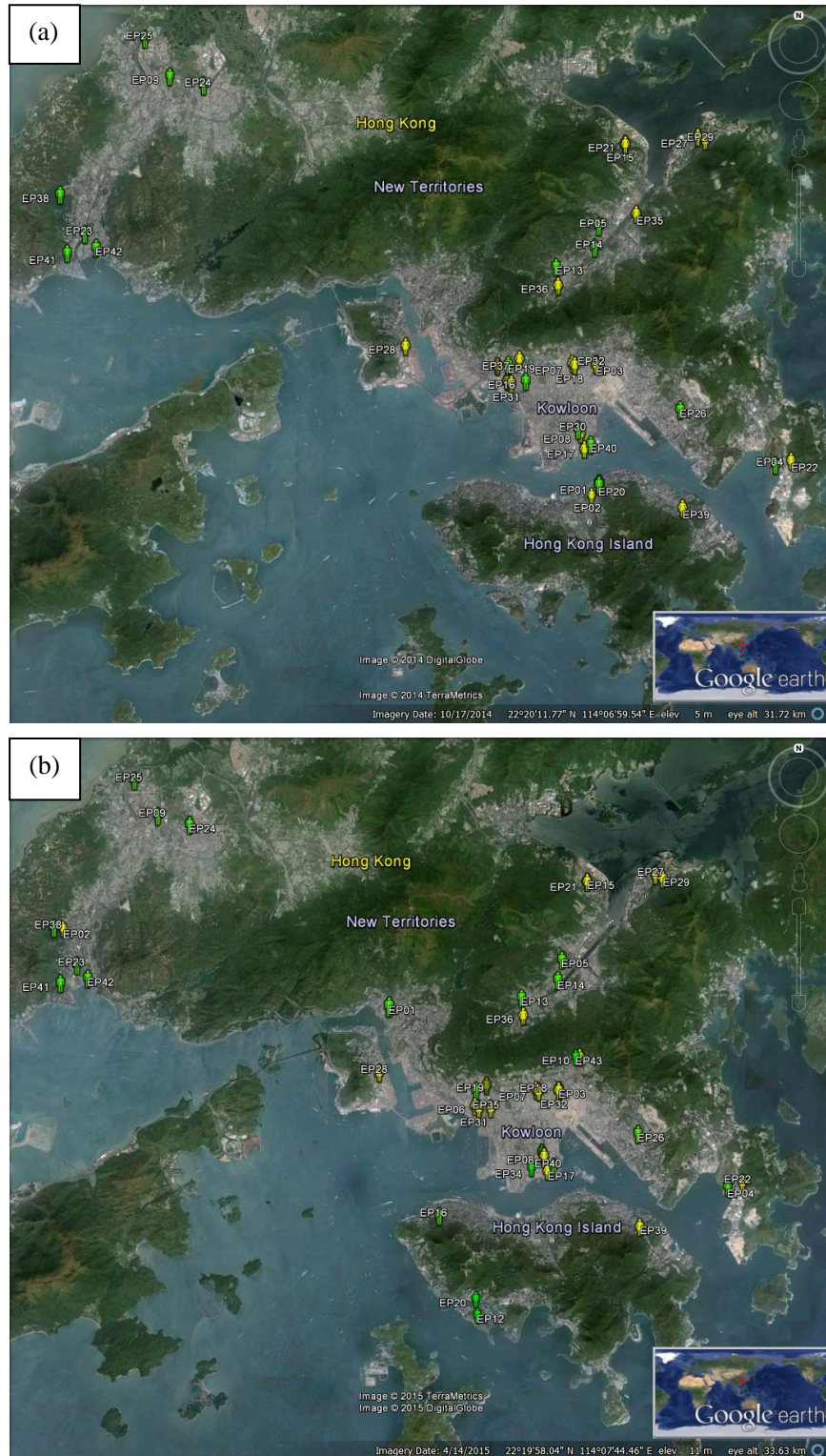
Figure 2-1. Map for Locations of the Subjects' Home Involved in This Study During Summer (a) and Winter (b) Campaign

Schedule B

The remaining twelve (12) recruited subjects were separated into three (3) groups

and personal PM$_{2.5}$ samples were collected from each subject in the once-every-18-days sampling frequency during the one year-long campaign. The participants' resident locations in different areas are shown in Figure 2-2. The personal PM$_{2.5}$ sampling date matched with the ambient PM$_{2.5}$ sampling program recommended by the HKEPD. The tentative sampling schedule from April 2014 to June 2015 is listed in Appendix D for reference. More than two hundred and forty (240) personal exposure samples were collected during April 2014 and June 2015.
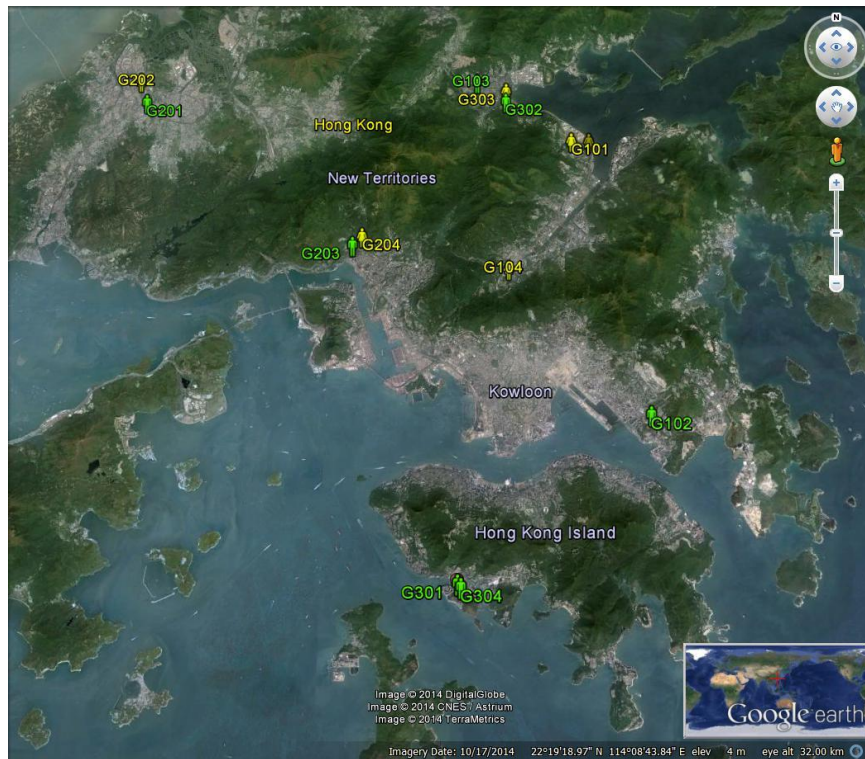
Figure 2-2. Map for Locations of the Subjects' Home Involved in This Study in Hong Kong for Schedule B

Information collected in questionnaires related to the subjects, their activities performed, journeys carried out, microenvironment visited, ventilation patterns and indoor sources have been collected to support the personal exposure sampling data.

Personal exposure to PM$_{2.5}$ was collected by a PM$_{2.5}$ Personal Environmental Sampler at a flow rate of 10 L/min. At this flow rate, a volume of 14.4 m^3 of air would be sampled for a 24-hr sampling period, and the starting time was set at midnight before transferring to the participant. The Personal Environmental Sampler consists of a sample inlet, namely Personal Environmental Monitor (PEM, model 761-203B; SKC Inc, PA,

USA, which is comprised of a red 2.5-µm nozzle cap, an impaction ring assembly and a base with 37 mm after filter), a battery-powered personal Leland Legacy Pump (SKC Inc., PA, USA) and a tube connecting the inlet and pump. A sampler power charger was provided to each participant for re-charge service during the evening period. Standard Operation Procedure (SOP) for Personal Sampling is attached in Appendix E. The PEMs were carried by the subjects at all times except sleeping, sitting and bathing during the 24-hr sampling period. The PEMs' inlets were re-examined near the breathing zone of each subject to mimic actual personal exposure. Individuals participating in the Schedule A of this study were outfitted with a backpack containing two pumps connected with two impactors. One Personal Environmental Monitor (PEM) loaded with a Teflon filter (37 mm, 2 µm pore size, Pall Teflon, Pall Corporation, Ann Arbor, MI, USA) and another one loaded with a quartz fiber filter (37 mm, Pall Tissuquartz Filter, Pall Corporation, Ann Arbor, MI, USA) was carried simultaneously by each subject on each sampling day. Individuals participate in Schedule B of this study carried a pump connected to an impactor with a 37 mm diameter Teflon-membrane filter only for one sampling day (Figure 2-3).

Figure 2-3. Personal Sampling Equipment

2.2.5 *Acceptance Test and Inter-Comparison Studies*

Before initial Personal Environmental Samplers operation, an acceptance test was carried out to confirm the samplers provided by Contractor were in good condition. Inter-

comparison studies of Personal Environmental Sampler were conducted at The Hong Kong Polytechnic University (PolyU) by Partisol Ambient Particulate Samplers in a trial period to demonstrate that both Personal Environmental Samplers provided by the Contractor could enhance data comparability and reliability.

2.2.6 *Preliminary Test*

Before official field sampling, a preliminary test was conducted: (1) To ensure the Personal Environmental Sampler was operated at an optimum level; (2) To ensure the operational procedure of personal exposure sampling was applied the maximum condition. The preliminary test procedure was listed as follows: (1) Preliminary test was performed on 1st of April 2014. Three field samples and three field blank samples were collected at each sampling location; (2) Three collocated samples were collected at the PolyU on the 3-4th of April 2014 in order to evaluate the deviation between Personal Environmental Samplers; (3) The sampling and analytical methods for filter samples were same as the routine monitoring protocol that provided at following sections.

2.2.7 *Time-Activity Diary*

During the personal exposure sampling period, each participant was required to keep a thirty-minutes interval time-activities diary, recording the time they spent in microenvironments (e.g., residential indoor, workplace, other buildings) in addition to information about exposure to environmental tobacco smoke (frequency and duration) and other potential exposures (e.g., use of cleaning products, house decoration). Further details of ventilation parameters in an enclosed environment were recorded. The indoor characters of sampling locations such as possible sources related to re-decoration and smoking were also recorded. The Chinese version of the time-activity diary was given to non-native English speaking participants. A time-activity diary template is attached in Appendix F.

2.2.8 *Sample Preparation and Analysis*

Filter preparation and Gravimetric analysis were conducted in a high-efficiency particulate absorption (HEPA) clean room that satisfied ISO 14644 Class 7 at The Hong

Kong Polytechnic University. Two types of 37 mm diameter filters (i.e. Quartz fiber filter, Teflon-membrane filter) were prepared in this study. Microbalance and working mass standards were calibrated by external contractor annually. Temperature (T), relative humidity (RH) and working mass standards verification was calibrated in every quarter. The balance was calibrated with 200 and 100 Class 1 standard weights and tare was set before weighing each batch of filters. After weighing every 10 filters, 200 and 100 mg calibration and tare were re-applied. All initial filters weighing was carried out 30 days before the sampling period. Post-sampling weighing was carried out no later than 30 days after the end of the sampling period.

Quartz fiber filters were pre-fired (900˚C, 3-hr) before sample collection to remove any potential contaminants on the filters. The particle mass on filter substrates was determined gravimetrically by weighing on an electronic microbalance with ± 1 μg sensitivity (Model MC-5; Sartorius AG, Goettingen, Germany) on a vibration-free working platform. The weighing conditions were controlled by recording the temperature and relative humidity before and after each session. All pre-/post- sampling filters, including quartz and Teflon filters, were transferred to a desiccator for 24-hr with a controlled temperature between 20-23 ± 2 ˚C and relative humidity of 30-40 ± 5% before weighing process. The charge on each filter was neutralized by exposure to a ^{210}Po ionizing source with no less than 500 picocuries for 30 seconds before filter weighting. Clean filters (Teflon and quartz filters) were stored in the drying box (RH < 40%) before sample collection. All samples and filter blanks were labeled immediately and preserved. After the sample collection, all sample filters were stored in Petri dishes, and stored in a freezer at -20 ˚C until further chemical analysis to prevent any evaporation of volatile components. Standard Operation Procedure (SOP) for Filter Weighing is attached in Appendix G. Sample details were filled in a sampling log sheet and then sent to the laboratory after verification by the project manager. A standard sampling log sheet is attached in Appendix H. Pre-sampling record was completed by laboratory staffs prior transferring to the Personal Environmental Samplers and finally to the subjects. Information on the sampling log sheet is with signatures and dates of all custodians:

1	Identification serial number of person conducting sampling

2	Subject information
3	Sampling location, date, and time
4	Filter IDs
5	Sample IDs
6	Sample flow rates (Leland Pump, Initial and Final flow rate from DryCal)
7	Total sample time
8	Total sample volume

The samples were collected by the person-in-charge at the laboratory. A duplicate record was kept.

2.2.9 ***Contingency Plan***

The sampling period of each subject was 24-hr, and a sampling program was set in sampler before the sampling period.

Samples quality was maintained by transferring the Personal Environmental Monitor to subjects 12-hr (i.e. not more than 24-hr) before sampling period. The purpose of establishing a contingency plan is to ensure research staff safety and subjects in the meantime preserving data integrity.

Subjects Participate in Schedule A

The sampling arrangement of subjects in Schedule A accommodates weather conditions. For example, re-arrangement of sampling schedule is essential if it is likely to rain, downpour, typhoon or any extreme weather conditions. The sampling is postponed, and subjects are immediately informed and requested for re-sampling on the next day if the weather condition is acceptable.

Subjects Participate in Schedule B

Subjects participate in Schedule B conducts sampling for all weather conditions except typhoon No. 8, or red/black rainstorm signal is hoisted. If the Personal Exposure Monitor is in malfunction during sampling, the subject is requested to conduct re-sampling, which follows the sampling schedule in next group. For example, a subject in

Group A failed to collect a sample in one day during the week (or during the weekend), he/she is requested to conduct re-sampling in Group B or Group C on coming scheduled weekday (or scheduled weekend).

2.3 Sample Analysis

All of the sample filters were analyzed by in laboratories, and a randomly selected ten percent (10%) of the samples were subjected to repeated analysis in external laboratories and internal laboratories for QA/QC purpose. The arrangement can be seen in the following Table 2-1.

Table 2-1. Analytical Methods for the Personal Exposure to Particulate Matter Study in Hong Kong

Filter Types	Analytical Methods	Laboratories (All samples)	Performance test (10% repeated)
Teflon	Gravimetry (Mass)	HKPU	HKPU
	X-Ray Fluorescence (40 elements from Na to U)	DRI	DRI
Quartz	Gravimetry	HKPU	HKPU
	Thermal/Optical Reflectance or Transmission (Organic Carbon and Elemental Carbon, OC/EC)	IEECAS	DRI
	Ion Chromatography (Cl^-, NO_3^- and SO_4^{2-}, Na^+, NH_4^+, K^+, Mg^{2+} and Ca^{2+})	HKBU	HKBU&HKPU

2.3.1 *Chemical Characterization*

As shown in Table 2-1, the Teflon-membrane filter collected samples for gravimetric analysis and elemental analysis. The Quartz fiber filter was analyzed for mass by gravimetry, for water-soluble anions (Cl^-, NO_3^-, and SO_4^{2-}) and cations (Na^+, NH_4^+, K^+, Mg^{2+}, and $Ca^{2+)}$ by ion chromatography, and for carbonaceous aerosols by thermal evolution method.

Elemental analysis

The post-sampled Teflon filters were shipped to Desert Research Institute (DRI) in temperature-controlled package (< 4 ˚C) to conduct elemental analysis (40 elements, including Na, Mg, Al, Si, P, S, Cl, K, Ca, Ti, V, Cr, Mn, Fe, Co, Ni, Cu, Zn, Ga, As, Se, Br, Rb, Sr, Y, Zr, Mo, Pd, Ag, Cd, In, Sn, Sb, Ba, La, Au, Hg, Tl, Pb, and U) by Energy Dispersive X-Ray Fluorescence (ED-XRF) spectrometry (PANalytical Epsilon 5, Almelo,

The Netherlands) (Chow and Watson, 2012).

Water-soluble inorganic ions

The quartz-fiber filters were analyzed for inorganic ions (anions: chloride, nitrate, sulfate (Cl^-, NO_3^- and SO_4^{2-}, respectively); and five cations sodium, ammonium, potassium, calcium (Na^+, NH_4^+, K^+, Mg^{2+} and Ca^{2+})) using a Dionex ICS-3000 ion Chromatograph (IC) at Hong Kong Baptist University (HKBU). A fixed area (2.63 cm^2) of the loaded filter sample was extracted with 4.0 mL of distilled–deionized water (DDI) under ultrasonic agitation for 120 min. The water extracted solutions were filtered through a syringe filter (0.45 μm, Pall Corporation, NY, USA) to remove insoluble material. All filtered solutions were stored at four ˚C until IC analysis (Ho et al., 2014).

Organic Carbon and Elemental Carbon analyzes

Organic carbon (OC) and elemental carbon (EC) were analyzed for each quartz filter samples from a 0.5 cm^2 punch using a Desert Research Institute (DRI) Model 2001 Thermal/Optical Carbon Analyzer following the IMPROVE_A thermal/optical reflectance (TOR) protocol (Cao et al., 2003; Chow et al., 2011). The protocol provided data following four OC fractions (OC1, OC2, OC3 and OC4 in a non-oxidizing helium atmosphere at 140˚C, 280˚C, 480˚C, and 580 ˚C), one OPC fraction (a pyrolyzed carbon) and three EC fractions (EC1, EC2 and EC3 at temperatures of 580˚C, 740˚C, and 840˚C in an oxidizing atmosphere of 2% oxygen in a balance of 98% helium). The amount of carbon measured after oxygen was added until the reflectance achieves its original value was reported as optically detected pyrolyzed carbon (OPC). The eight fractions OC1, OC2, OC3, OC4, EC1, EC2, EC3, and OPC were reported separately in the data sheet. IMPROVE_A OC was operationally defined as OC1 + OC2 + OC3 + OC4 + OPC, whereas EC was defined as EC1 + EC2 + EC3 − OPC. The detection limits for OC and EC were below 1.0 μg m^{-3} (0.28 μg m^{-3} for OC and 0.04 μg m^{-3} for EC).

2.3.2 *Instrument Calibration and Frequency*

Flow check was conducted by connecting a Personal Environmental Sampler to a DryCal® DC-Lite Primary Air Flow Meter (Model DCL-MH; Bios International Corp.,

Butler, NJ, USA). Calibration Cap (Model 761-202; SKC Inc, PA, USA) was attached to a filter loaded PEM, and the inlet of the Calibration Cap was connected to the flow meter for flow checking and calibration. A routine flow check of the Personal Environmental Samplers was conducted before and after each sampling. The recommended flow rate for the PEM specified in this study was 10 L/min. Start the pump, adjust the flow to 10 L/min, and run the pump for a minimum of 5 min to stabilize the flow. Adjust pump flow until 10 L/min appears on the flowmeter, and record the flow rate. At the end of the sampling period, document the final flow rate displayed on the flowmeter. The difference between initial and final flow rate varied from sample to sample but was typically within ± 5% of the pre-set volume.

2.3.3 *Quality Assurance and Quality Control (QA/QC)*

Appropriate quality assurance/quality control (QA/QC) were implemented in the handling of filters, personal sampling, IMPROVE_TOR, IC and XRF to the satisfaction of EPD to ensure data quality. Standard Operation Procedures for these analysis were attached in respective Appendix.

A preliminary test was conducted before the official sampling campaign on the 3rd-4th of April 2014 to ensure the PEMs were in suitable condition by using two sets (three collocated samples) of 37 mm diameter filters (Teflon and quartz) to collect $PM_{2.5}$ in the PEM at The Hong Kong Polytechnic University (HKPU). Ambient $PM_{2.5}$ samples were simultaneously collected by two Mini-vol samplers (Airmetrics, Eugene, OR, USA) with each loaded with a 47 mm quartz fiber filter and one 47 mm Teflon filter respectively. The 37 mm filters were pre-fired in the same manner as the 47 mm filters. As shown in Table 2-4, in the side-by-side pilot test for PEMs and Mini-vol sampler, the average personal $PM_{2.5}$ values (7.6 ± 1.0 µg m^{-3}) loaded with quartz filters showed comparable value with the ambient $PM_{2.5}$ sample (7.6 µg m^{-3}). The deviation of the PEMs loaded with quartz filters expressed in coefficient of variance (CV (%)) was 13.6%. The average personal $PM_{2.5}$ mass concentration (7.6 ± 0.9 µg m^{-3}) of three collocated Teflon filter samples was higher than the ambient $PM_{2.5}$ concentrations (6.7 µg m^{-3}). The coefficient variance of these three PEMs was 11.4%.
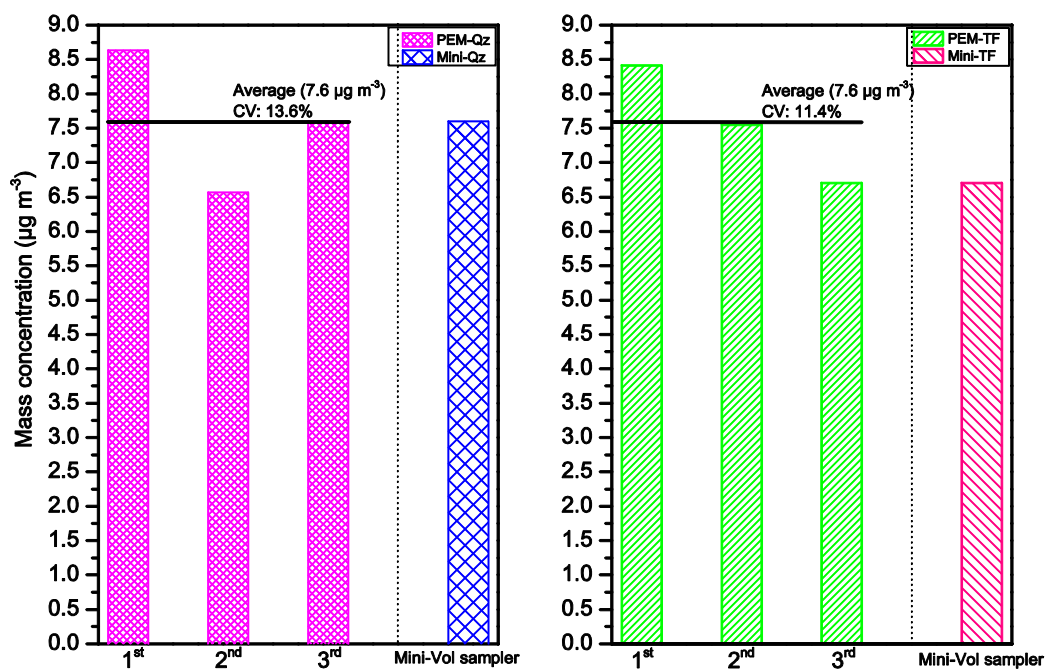
Figure 2-4. Inter-comparison of PEM$_S$

In general, ten percent (10%) of the collected samples was subjected to repeated gravimetric analysis for QA/QC purposes. As illustrated in Figure 2-5, 10 and 11 Teflon filter samples were selected and the second post-weighing procedure was conducted during summer and winter sampling campaign, respectively. The mass difference of the first and second average final weights was in a range of ± 1.5 µg m^{-3}. As shown in Figure 2-6, 9.2% of the Teflon filter samples were selected and subjected to repeated gravimetric analysis, and the mass difference between the first and second average final weights of the sample was within ± 1.0 µg m^{-3}. It can also be seen that the absolute difference (%) between the post-weights were within ± 15% (Figure 2-5 and 2-6).
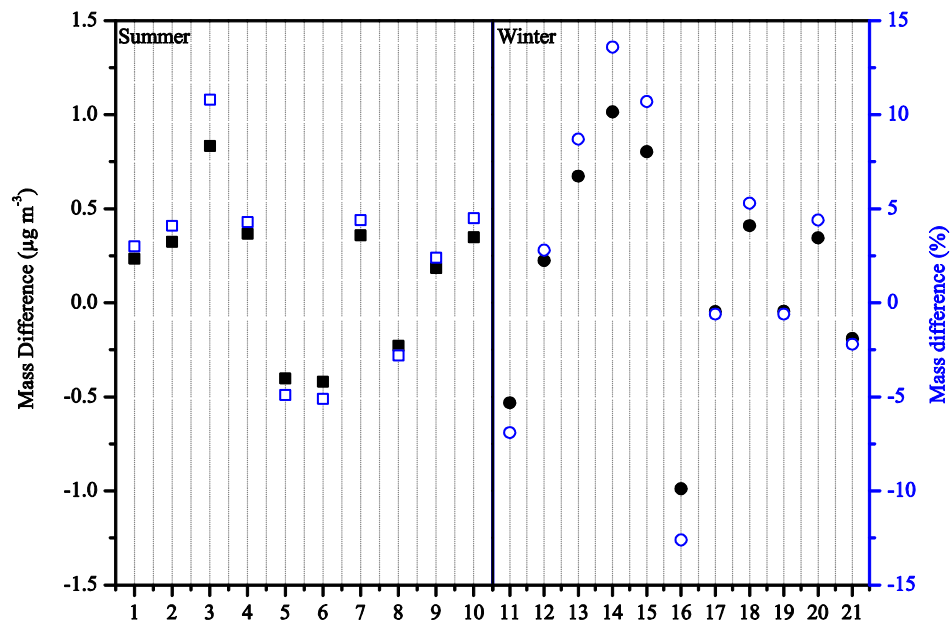
Figure 2-5.Mass Difference of the Replicate Post-weighing During Summer and Winter Campaign of Schedule A
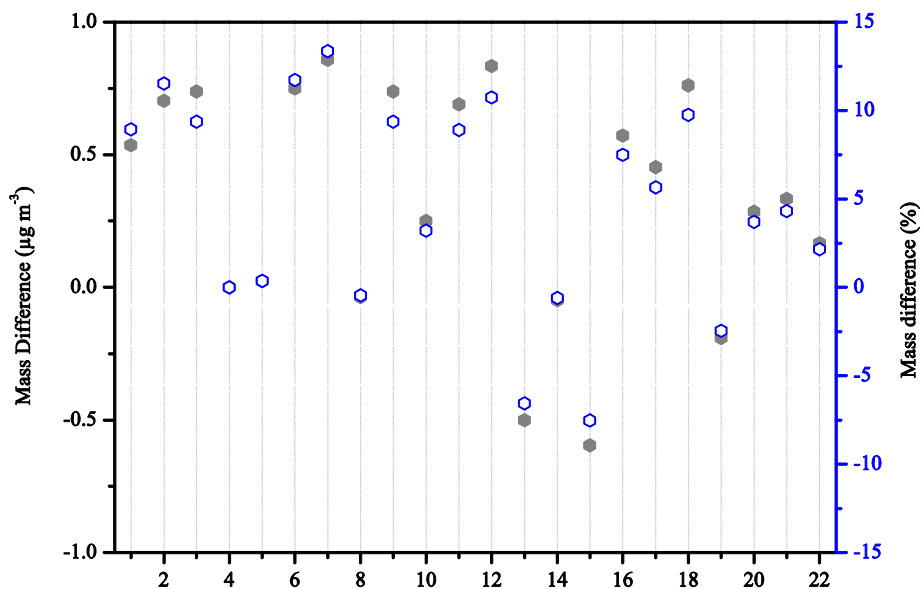
Figure 2-6. Mass Difference of the Duplicate Post-weighing of Schedule B

Table 2-2. Comparability Measures for OC, EC and TC on DRI Model 200l Carbon Analyzer at DRI and IEECAS

Laboratory	IMPROVE_TOR	Sample ID		
		A (μg C cm^{-2})	B (μg C cm^{-2})	C (μg C cm^{-2})
DRI	OCTRC	38.70	54.89	10.35
	ECTRC	10.95	14.79	2.74
	TCTC	49.64	69.68	13.09
IEECAS	OCTRC	41.45	53.12	10.72
	ECTRC	9.59	13.96	2.54
	TCTC	51.04	67.08	13.26
Mass Difference (%)	OCTRC	-6.9%	3.3%	-3.5%
	ECTRC	13.2%	5.8%	7.6% (0.20 < 1.0)
	TCTC	-2.8%	3.8%	-1.3%

The comparability and equivalence of analysis methods were established by the comparison of the results for the same measurement (IMPROVE_TOR), and intra-laboratory comparison (DRI&IEECAS) were made to establish the comparability for OC/EC in this study. As shown in Table 2-2, the carbon fractions compare well between the two laboratories.

Table 2-3. Comparability Measures for Anions Standard at HKBU and HKPU

Laboratory	IC (Standard)	Chemical species		
		Cl$^-$ (mg/L)	NO$_3^-$ (mg/L)	SO$_4^{2-}$ (mg/L)
HKBU	0.5	0.604	0.566	0.578
	1.0	1.115	1.055	0.941
	2.0	1.863	1.944	1.944
	4.0	3.608	3.805	3.721
	10.0	9.993	9.947	9.973
HKPU	0.5	0.508	0.554	0.526
	1.0	1.028	1.024	0.994
	2.0	2.100	2.199	2.185
	4.0	3.851	4.024	3.907
	10.0	10.036	9.945	9.989
Precision (%)	0.5	6.8%	0.8%	3.7%
	1.0	6.1%	2.1%	3.7%
	2.0	16.7%	18.0%	17.1%
	4.0	17.2%	15.5%	13.2%
	10.0	3.1%	0.1%	1.1%

The precision of ions analysis has been shown in Table 2-3, and the precision of Cl$^-$, NO$_3^-$, and SO$_4^{2-}$ have been reported to range from 3.1%-17.2%, 0.1-18.0%, and 1.1-

17.1%, respectively. The precision of anion analysis compares well ($< 20\%$) between the two laboratories (HKBU&HKPU).

Table 2-4. Ions Replicates in Personal $PM_{2.5}$ Samples

Sample ID	Cations ($\mu g\ m^{-3}$)					Anions ($\mu g\ m^{-3}$)		
	Na^+	NH_4^+	K^+	Ca^{2+}	Mg^{2+}	Cl^-	NO_3^-	SO_4^{2-}
20141011EP06	-1.0%	-0.4%	-1.0%	-7.5%	-7.0%	2.2%	3.4%	3.5%
20140817EP12	-0.4%	-1.7%	-4.3%	-16.5%	1.2%	18.3%	6.6%	4.3%
20140912EP17	0.4%	-1.8%	-0.3%	5.1%	-1.9%	1.1%	5.0%	0.0%
20140917EP22	-2.3%	-1.4%	-1.7%	-6.6%	-0.7%	-1.2%	-1.7%	-1.2%
20140926EP28	-1.4%	-0.6%	0.3%	3.6%	-1.9%	-3.4%	-1.4%	1.9%
20141012EP34	-3.4%	-1.2%	-1.3%	-2.7%	-5.2%	-5.5%	-4.4%	-0.2%
20141013EP39	4.4%	3.5%	2.1%	-3.4%	1.6%	0.2%	-2.3%	-2.0%
20150302EP04	-0.8%	-0.8%	-1.9%	-10.9%	-6.9%	6.9%	6.8%	5.9%
20150108EP07	8.1%	4.3%	6.0%	3.8%	2.0%	-1.8%	-0.8%	0.4%
20150313EP10	0.5%	0.1%	1.1%	-1.5%	7.4%	-0.8%	-1.3%	-1.5%
20141217EP14	-2.6%	-3.2%	-2.1%	0.8%	-0.4%	0.6%	-1.1%	5.5%
20141231EP17	-1.0%	-0.8%	-1.1%	-3.4%	4.7%	-2.4%	-0.5%	-1.0%
20150203EP19	2.0%	1.4%	1.6%	-1.5%	-0.9%	10.0%	11.2%	9.4%
20150226EP25	1.2%	0.7%	2.3%	-2.0%	12.6%	-2.5%	2.3%	3.3%
20150205EP31	5.5%	2.2%	3.5%	3.0%	9.8%	2.4%	4.0%	4.4%
20150302EP38	6.0%	3.4%	4.6%	3.1%	7.1%	0.5%	4.4%	5.1%
20150119EP41	5.4%	2.3%	3.5%	8.7%	11.2%	-2.8%	-0.3%	-0.4%
Mean	2.7%	1.7%	2.3%	5.0%	4.9%	3.7%	3.4%	2.9%
Standard Deviation	2.3%	1.2%	1.6%	4.1%	3.9%	4.5%	2.9%	2.6%

Replicates of analyzed samples were randomly selected and performed for every ten samples (N =17). As illustrated in Table 2-4, compare replicates against the original run, and no differences were found that exceeded allowable tolerance (Watson et al., 2001). The absolute average difference between main and replicate results for Cl^-, NO_3^-, SO_4^{2-}, Na^+, NH_4^+, K^+, Mg^{2+} and Ca^{2+} in personal $PM_{2.5}$ were $3.7 \pm 4.5\%$, $3.4 \pm 2.9\%$, $2.9 \pm 2.6\%$, $2.7 \pm 2.3\%$, $1.7 \pm 1.2\%$, $2.3 \pm 1.6\%$, $5.0 \pm 4.1\%$, $4.9 \pm 3.9\%$, respectively.

2.3.4 *Analytical Specification*

The sampling dates on which the samples were collected during Schedule A and Schedule B is listed in Appendix D. A total of 172 sets (Teflon and quartz filter) of 24-hr personal $PM_{2.5}$ samples were collected during summer (90) and winter (82). A total of 254 Teflon filter samples was collected in Schedule B. Eleven samples were identified to be invalid during the study period. The corresponding sample IDs, filter IDs, subject IDs,

and calculated mass concentrations were listed in Table 2-5. In summary, 167 out 172 and 248 out of 254 personal $PM_{2.5}$ samples collected in Schedule A and Schedule B, respectively, were considered valid after data validation and final review. This resulted in more than 12000 data points, as documented in Section 3.

Table 2-6 summarizes the analytical specifications for the 24-hr personal $PM_{2.5}$ samples obtained during the whole study period. The number of reported (valid) values for each chemical species and the number greater than the Method detection limit (MDL) were also summarized in Table 2-5. Mass concentrations (both Teflon and quartz), ions (including chloride, nitrate, sulfate, ammonium, soluble sodium, soluble potassium, soluble calcium, and soluble magnesium), organic carbon and elemental carbon, sodium (Na), aluminum (Al), silicon (Si), sulfur (S), chlorine (Cl), potassium (K), calcium (Ca), titanium (Ti), vanadium (V), iron (Fe), nickel (Ni), copper (Cu), zinc (Zn), bromine (Br) were detected in almost all samples (more than 90%), especially crustal materials (e.g., Al, Si, Ca, Ti, Fe) were detected in 95% of the samples. Magnesium (Mg) was detected in 95.8% and 96.7% of the personal $PM_{2.5}$ samples in Schedule A and Schedule B, respectively. Several transition metals (e.g., Co, Pd, Ag, Cd, In, Sb, Ba, La, Hf, Ta, Ir, Au, Hg, Tl) were not detected in most of the personal samples (less than 5%). This is typical for ambient samples collected at urban and non-urban sites. Arsenic (As) was found above the MDL in 70.9% and 65.0% of the analyzed personal samples, respectively. Motor vehicle exhaust species (e.g., Br, Pb) were detected in 98.2%-99.2% and 88.2%-89.9% of the personal $PM_{2.5}$ samples. Residual-oil-related species (e.g., V, Ni) were detected in 97.0% and 96.7% of the personal samples in Schedule A and in 98.0% and 100% of personal samples in Schedule B. Industrial-source-related, toxic species (e.g., Cd, Hg) were not detected (0%) in personal samples.

The analytical specifications (Table 2-6) indicated that personal $PM_{2.5}$ samples collected during the study period possess adequate sample loading for chemical analysis. The method detection limits of the listed analytical methods were sufficiently low to establish valid measurements with acceptable precisions.

Table 2-5. List of Invalid Samples (Tender Reference No. 13-04909)

	Sample ID	Filter ID	Subject ID	PM concentration ($\mu g\ m^{-3}$)	Remarks
Schedule A	HKEPD20141014	EPDFT061	EP21	24.0	Chemical data showed that the weighing result of Teflon filter was too low and there was large discrepancy between filter weights and chemical concentrations (OC = 9.1 $\mu g\ m^{-3}$; EC = 2.6 $\mu g\ m^{-3}$; NH_4^+ = 13.1 $\mu g\ m^{-3}$; SO_4^{2-} = 34.3 $\mu g\ m^{-3}$)
	HKEPD20141013	EPDFT062	EP39	55.2	Abnormally high OC concentration (OC = 42.7 $\mu g\ m^{-3}$, EC = 1.9 $\mu g\ m^{-3}$)
	HKEPD20140902	EPD20140717080	EP07	102.7	Chemical data showed that the weighing result of Teflon filter was too high, and there was large discrepancy between Qz filter weight (17.5 $\mu g\ m^{-3}$) and Tf filter weight
	HKEPD20150105	EPDFT090	EP30	107.4	Chemical data showed that the weighing result of Teflon filter was too high, and there was large discrepancy between Qz filter weight (54.3 $\mu g\ m^{-3}$) and Tf filter weight
	HKEPD20150119	EPDFT109	EP36	55.5	Chemical data showed that the weighing result of Teflon filter was too low, and there was large discrepancy between filter weights (Qz concentration = 111.5 $\mu g\ m^{-3}$)
Schedule B	HKEPD20140513	EPDST1403122	G204	74.4	Total sample time (398 min) less than 16-hr
	HKEPD20140519	EPDST1403124	G301	38.6	Total sample time (524 min) less than 16-hr
	HKEPD20140606	EPDST1403140	G304	52.4	Total sample time (680 min) less than 16-hr
	HKEPD20140624	EPD140530052	G302	N.A.	Total sample time was not available due to pump malfunction
	HKEPD20141215	EPD20140707ST013	G204	N.A.	Total sample time was not available due to pump malfunction
	HKEPD20150201	EPDFT115	G103	N.A.	Negative mass concentration

Table 2-6. Analytical Specifications for 24-hr Ambient and Personal $PM_{2.5}$ Measurements During the Personal Exposure to Particulate Matter Study in Hong Kong

Species	Analysis Methods	MDL^b ($\mu g\ m^{-3}$)	Blank Subtracted^c	Schedule A			Schedule B (Including ambient samples)		
				N^d	No. > MDL	% > MDL	N	No. > MDL	% > MDL (Personal sample)
Teflon Mass	Gravimetry	N.A.	0.00	167	N.A.	N.A.	760	N.A.	N.A.
Quartz Mass	Gravimetry	N.A.	0.00	141	N.A.	N.A.	310	N.A.	N.A.
Chloride (Cl^-)	IC	0.03	0.33 (mg/L)	167	162	96.4%	512	438	85.5%
Nitrate (NO_3^-)	IC	0.01	0.00 (mg/L)	167	163	97.0%	512	501	97.9%
Sulfate (SO_4^{2-})	IC	0.01	0.05 (mg/L)	167	167	100.0%	512	468	91.4%
Ammonium (NH_4^+)	IC	0.23	0.08 (mg/L)	167	128	76.8%	N.A.	N.A.	N.A.
Soluble Sodium (Na^+)	IC	0.18	0.03 (mg/L)	167	163	97.6%	N.A.	N.A.	N.A.
Soluble Potassium (K^+)	IC	0.01	0.06 (mg/L)	167	165	98.8%	512	388	75.8%
Soluble Calcium (Ca^{2+})	IC	0.04	0.15 (mg/L)	167	167	100.0%	512	496	96.9%
Soluble Magnesium (Mg^{2+})	IC	0.02	0.58 (mg/L)	167	167	100.0%	512	512	100.0%
IMPROVE_A									
OCTC	TOR	0.28	1.60 ($\mu g\ C\ cm^{-2}$)	167	167	100.0%	512	505	98.6%
ECTC	TOR	0.04	0.11 ($\mu g\ C\ cm^{-2}$)	167	167	100.0%	512	509	99.4%
TCTC	TOR	0.28	1.63 ($\mu g\ C\ cm^{-2}$)	167	167	100.0%	512	505	98.6%
Sodium (Na)	XRF	0.0331	1.4454 (μg /filter)	162	161	100.0%	758	756	99.7% (100%)
Magesium (Mg)	XRF	0.0120	0.0000 (μg /filter)	117	112	95.8%	693	548	79.1% (96.7%)
Aluminium (Al)	XRF	0.0048	0.0499 (μg /filter)	156	153	98.1%	751	704	93.7% (97.9%)
Silicon (Si)	XRF	0.0030	0.0193 (μg /filter)	164	162	98.8%	750	717	95.6% (98.7%)
Phosphorus (P)	XRF	0.0027	0.0000 (μg /filter)	N.A.	N.A.	N.A.	514	339	66.0% (100%)
Sulfur (S)	XRF	0.0024	0.0021 (μg /filter)	167	167	100.0%	760	760	100.0% (100%)
Chlorine (Cl)	XRF	0.0048	0.0040 (μg /filter)	167	162	97.0%	760	723	95.1% (98.8%)
Potassium (K)	XRF	0.0029	0.0033 (μg /filter)	167	167	100.0%	760	760	100.0% (100%)
Calcium (Ca)	XRF	0.0022	0.0169 (μg /filter)	166	166	100.0%	760	760	100.0% (100%)
Titanium (Ti)	XRF	0.0014	0.0000 (μg /filter)	165	162	98.2%	756	698	92.3% (98.0%)
Vanadium (V)	XRF	0.0012	0.0000 (μg /filter)	167	162	97.0%	757	747	98.7% (98.0%)
Chromium (Cr)	XRF	0.0009	0.0000 (μg /filter)	119	86	72.5%	730	537	73.6% (85.8%)
Manganese (Mn)	XRF	0.0008	0.0000 (μg /filter)	161	157	97.5%	752	669	89.0% (94.6%)
Iron (Fe)	XRF	0.0007	0.0000 (μg /filter)	167	167	100.0%	760	760	100.0% (100%)
Cobalt (Co)	XRF	0.0004	0.0000 (μg /filter)	3	0	0.0%	521	140	26.9% (0%)
Nickel (Ni)	XRF	0.0004	0.0000 (μg /filter)	152	147	96.7%	751	736	98.0% (100%)
Copper (Cu)	XRF	0.0005	0.0000 (μg /filter)	161	160	99.4%	758	735	97.0% (98.8%)
Zinc (Zn)	XRF	0.0005	0.0000 (μg /filter)	167	167	100.0%	760	743	97.8% (100%)

Gallium (Ga)	XRF	0.0009	0.0000 (µg /filter)	11	0	0.0%	527	70	13.3% (0%)
Arsenic (As)	XRF	0.0008	0.0000 (µg /filter)	149	104	70.9%	718	329	45.8% (65.0%)
Selenium (Se)	XRF	0.0006	0.0000 (µg /filter)	121	75	62.3%	697	240	34.4% (64.3%)
Bromine (Br)	XRF	0.0005	0.0000 (µg /filter)	166	163	98.2%	760	721	94.9% (99.2%)
Rubidium (Rb)	XRF	0.0005	0.0000 (µg /filter)	145	104	71.9%	731	309	42.3% (71.2%)
Strontium (Sr)	XRF	0.0005	0.0000 (µg /filter)	166	139	83.8%	758	418	55.1% (90.7%)
Yttrium (Y)	XRF	0.0006	0.0000 (µg /filter)	91	33	37.0%	658	142	21.6% (39.0%)
Zerconium (Zr)	XRF	0.0008	0.0000 (µg /filter)	148	99	67.1%	724	268	37.0% (67.5%)
Molybdenum (Mo)	XRF	0.0013	0.0000 (µg /filter)	134	61	45.9%	720	251	34.9% (34.6%)
Palladium (Pd)	XRF	0.0053	0.0000 (µg /filter)	62	0	0.0%	587	5	0.9% (0%)
Silver (Ag)	XRF	0.0058	0.0000 (µg /filter)	23	0	0.0%	550	14	2.5% (0%)
Cadmium (Cd)	XRF	0.0058	0.0000 (µg /filter)	36	0	0.0%	577	11	1.9% (0%)
Indium (In)	XRF	0.0062	0.0000 (µg /filter)	52	0	0.0%	609	1	0.2% (0%)
Tin (Sn)	XRF	0.0081	0.0000 (µg /filter)	157	76	48.7%	726	179	24.7% (35%)
Antimony (Sb)	XRF	0.0086	0.0000 (µg /filter)	75	0	0.0%	613	20	3.3% (0%)
Barium (Ba)	XRF	0.0249	0.0000 (µg /filter)	22	10	47.8%	555	12	2.2% (0%)
Lanthanum (La)	XRF	0.0297	0.0000 (µg /filter)	43	31	72.7%	566	12	2.1% (0%)
Gold (Au)	XRF	0.0015	0.0000 (µg /filter)	54	1	1.8%	595	59	9.9% (2.4%)
Mercury (Hg)	XRF	0.0012	0.0000 (µg /filter)	6	0	0.0%	547	2	0.4% (0%)
Thallium (Tl)	XRF	0.0012	0.0000 (µg /filter)	12	0	0.0%	548	6	1.1% (0%)
Lead (Pb)	XRF	0.0014	0.0000 (µg/filter)	158	142	89.9%	750	612	81.6% (88.2%)
Uranium (U)	XRF	0.0011	0.0000 (µg /filter)	62	12	22.2%	601	38	6.3% (22.5%)

Notes: [a]IC = ion chromatography, XRF = X-ray fluorescence; [b]MDL refers to method detection limit; [c]Non-zero average blank concentration are subtracted; [d]N refers to the number of valid data; N.A. stands for data not available.

2.4   Data Management

The overall data management was to compose completed data set as soon as after data collection and a centralized data archive was established. The collected data were imported to the computer (e.g., excel format) for storage and delivered in documenting format on CD-ROM. Sample collection details (e.g., subject ID, sampling date, sampling time, particle size, sampling period) were manually input in software. These data were organized so that information about any species at any sampling locations could be readily identified and accessed. The role of project manager was to administrate data and identify also modify incorrect attributes in the collected data. All hard copies were for the reference. Project Manager was manually associate a "samples quality" flag with analysis results that were suspect due to sample collection or analytical problems.

2.5   Data Validation and Reporting

There are five types of common operations generating records which subject to data verification: 1) sample collection; 2) sample receipt; 3) sample preparation; 4) sample analysis; 5) records review.

Data verification process is done by the project manager to evaluate the completeness, correctness and compliance with a specific data set: 1) sample collection logs; 2) calculation worksheet; 3) calibration records; 4) quality control results against the method, procedure, or contractual requirements.

Data validation process was performed by project manager in order to: 1) evaluate the field records for consistency; 2) review QC information (e.g., calibration results, field blanks); 3) summarize deviations and determine impact on data quality; 4) summarize sample collection; 5) review summary of data verification to determine analytical method, procedural, and contractual required QC compliance/non-compliance; 6) review verified, reported sample results collectively for the dataset as a whole (e.g., calculation); 7) summarize data and QC deficiencies and evaluate the impact on overall data quality.

2.5.1   ***Sum of Chemical Species versus PM$_{2.5}$ Mass***

The sum of the chemical constituents measured in personal PM$_{2.5}$ should be less than or equal to the corresponding personal mass concentrations obtained from

gravimetric measurements. The chemical species included those that were quantified on both quartz filters and Teflon filters. Chloride (Cl⁻), total potassium (K), soluble sodium (Na⁺), and sulfate (SO_4^{2-}) were included, while sulfur (S), chlorine (Cl), total sodium (Na), total Magnesium (Mg) and potassium (K⁺) were excluded to avoid double counting. Organic carbon and elemental carbon concentrations were included in the summation of chemical species. Metal oxides, unmeasured ions, or hydrogen and oxygen associated with organic carbon were not included in the measured concentrations.
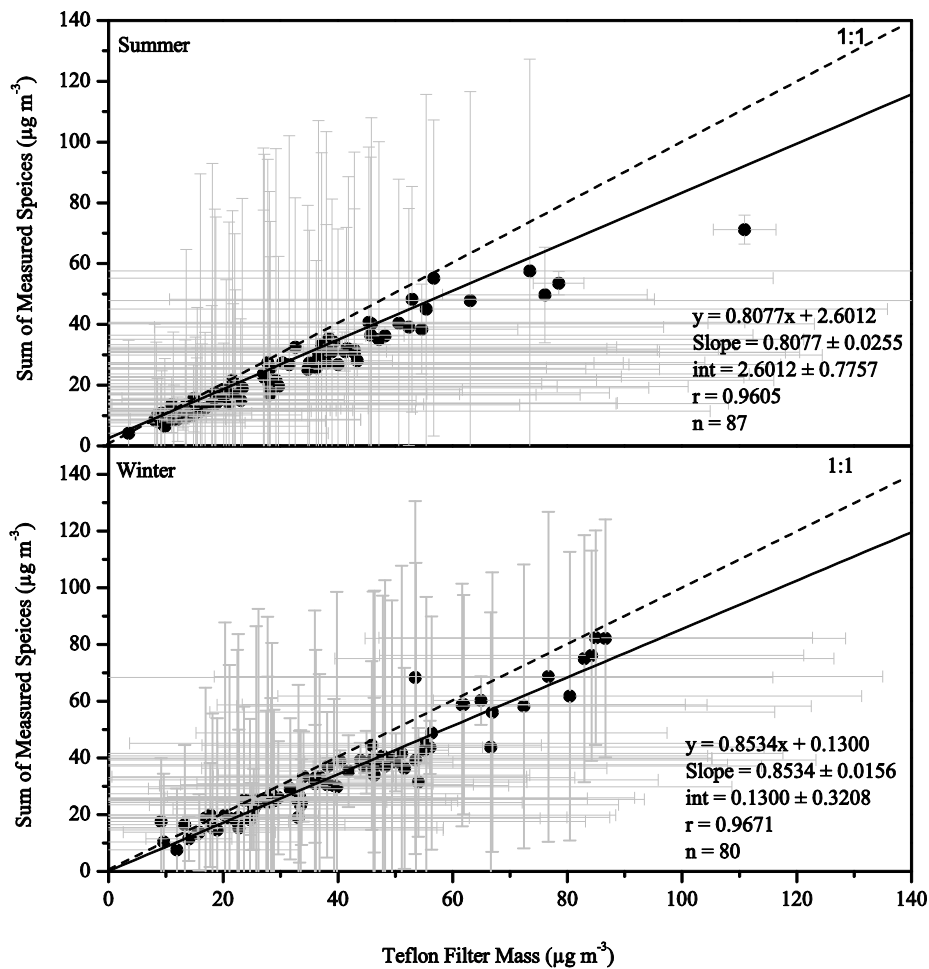
Figure 2-7. Scatter Plots of Sum of Chemical Species versus Measured Mass for Personal $PM_{2.5}$ Samples Collected During Summer and Winter

The sum of chemical species was plotted against with gravimetric personal $PM_{2.5}$

concentrations on Teflon filters obtained during summer and winter in Figure 2-7. Each plot contains a solid line indicating the slope with intercept and a dashed one-to-one line. Strong correlations (r: 0.96-0.97) were found between the sum of chemical species and mass concentrations with a slope of 0.81 ± 0.03 in summer and 0.85 ± 0.02 in winter, respectively. Approximately 83.1%-87.6% of the personal $PM_{2.5}$ mass was explained by the measured chemical species in this study. A good correspondence between the sum of chemical constituents and the measured mass indicates that a good analytical quality of individual chemical determinations is assured in this study.

2.5.2 *Reconstructed versus Measured Mass*

Major fine particle species can be classified into seven categories including: 1) geological material, which can be estimated as $1.89 \times [Al] + 2.14 \times [Si] + 1.4 \times [Ca] + 1.43 \times [Fe]$; 2) organic matter ($[OM] = 1.4 \times [OC]$); 3) soot (EC concentration); 4) inorganic ions including ammonium sulfate ($1.38 \times [SO_4^{2-}]$) and ammonium nitrate ($1.29 \times [NO_3^-]$; 5) sea salt (Na^+); 6) non-crustal trace elements; 7) unidentified material. Considering the large uncertainty in total sodium (Na) measurement by XRF, soluble sodium is used in the calculation instead of total sodium. The difference between constructed mass and the measured mass was referred as unidentified mass. The reconstructed personal $PM_{2.5}$ mass was calculated by the following equation:

[Reconstructed Mass] = $(1.89 \times [Al] + 2.14 \times [Si] + 1.4 \times [Ca] + 1.43 \times [Fe]) + (1.4 \times [OC] + [EC]) + (1.38 \times [SO_4^{2-}] + 1.29 \times [NO_3^-]) + [Na^+]$ + non-crustal elements excluding geological material (e.g, Al, Si, Ca, Fe, S)　　　　　　　　　　　　(2.1)
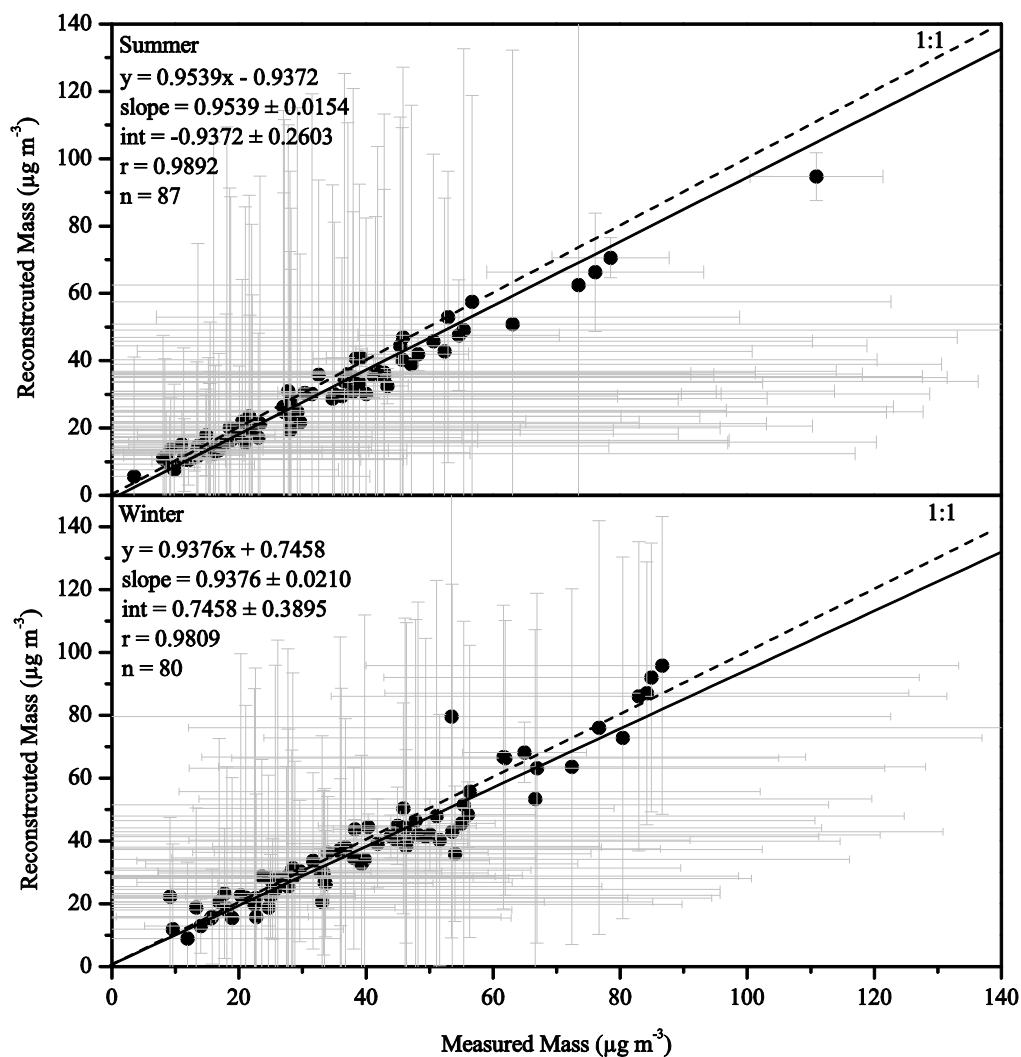
Figure 2-8. Scatter Plots of Reconstructed Mass versus Measured Mass for Personal PM$_{2.5}$ Data Acquired During Summer and Winter

The reconstructed mass was highly (r: 0.98-0.99) correlated with the measured mass, with a slope of 0.96 ± 0.02 and 0.94 ± 0.02 in summer and winter. In contrast with the sum of chemical species and measure mass in Figure 2-7, the major uncertainty of the reconstructed mass was due to the estimated of organic matter (OM).

3. RESULTS

3.1   Schedule A

3.1.1 _Study Group_

In summer, as shown in Table 3-1, the study group consisted of 20 female and 22 male subjects (approximately 1:1 gender ratio) between 18 and 63 years of age. Forty-one (41) subjects including 19 female and 22 male subjects participated in the winter campaign (Table 3-1). Seven subjects dropped out after the first sampling campaign, and six new subjects were subsequently recruited to participate in the second campaign. The remaining 35 subjects completed both the summer and winter campaign. All participants completed a detailed questionnaire about gender, age, occupation, home location, lifestyle factors, and workplace exposure. All subjects were categorized into four main groups: (1) students; (2) office workers; (3) housewives and (4) non-office workers.

Table 3-1. Description Detail of the Subjects During the Personal Sampling Campaign of Schedule A

	1st sampling (Summer)	2nd sampling (Winter)
Sampling Date	July 8 - October 2014	December 09, 2014- March 27, 2015
Study subjects (N*)	42 (35)	41 (35)
Female	20	19
Male	22	22
Median age, years (range)	28 (18-63)	27 (18-63)
Occupation (N)		
Students	14	16
Office workers	10	10
Housewives	10	10
Non-office workers	8	5
Smokers (N)	0	0

*Number of recruited subjects participated both in summer and winter campaign.

3.1.2 _Activity Profiles_

The subjects further completed time-activity diaries in a 24-hr monitoring period. The monitoring parameters included time at home, time at work, time outdoors and time spent on transportation (e.g., bus, minibus, metro). Time-activity diaries were completed every 30 min for each of the monitoring days. Ultimately, 82 and 79 activity diaries were collected during summer and winter campaign, respectively. According to the time-activity diaries of the subjects, a summary of time spent in different microenvironments associated with personal monitoring is shown in Table 3-2. Moreover, daily activity

patterns of subjects (including four main groups: students, office workers, housewives, and non-office workers) in Schedule A during the summer and winter were shown in Table S1-S4 (Appendix I). The subjects spent more than 90% of the sampling time in the indoor environment during summer. It was calculated that subjects spent 71.9% of their daily time at home and 13.6% at work (school) respectively. The daily time contribution of time spent on transportation and outdoor was equal to 5.7% and 2.4% respectively. As shown in Table 3-2, it was calculated that subjects spent 79.2% of their time at home, and 93.8% of their time in the indoor microenvironment during the second sampling period. A considerable portion of time was spent at work (13.2%) with the rest of the daily activities divided between transportation (4.0%) and outdoors (2.8%). Similar results were reported in the previous study in Hong Kong, on average, the subjects spent more than 86% of their time indoors, 3-7% in transit and 3-7% in outdoor (Chau et al., 2002).

Table 3-2. Subjects' Activity During the Personal Sampling Campaign of Schedule A

	1st sampling (Summer)	2nd sampling (Winter)
Time-activity data from diaries		
Time spent, median (%)		
Indoors, total	91.7%	93.8%
Indoors at home	71.9%	79.2%
Indoors at work (average)	13.6%	13.2%
Outdoors	2.4%	2.8%
Transportation (average)	5.7%	4.0%

3.1.3 *Personal PM$_{2.5}$ Exposure*

- Characterization of personal exposure to PM$_{2.5}$ mass concentrations

The descriptive statistics of the measured personal exposures in summer and winter were listed in Table 3-3. The average and median personal exposure to PM$_{2.5}$ concentrations obtained during the entire monitoring period were 34.9 ± 19.6 µg m^{-3} and 31.7 µg m^{-3} respectively. Personal PM$_{2.5}$ mass concentrations ranged from 3.5 to 110.9 µg m^{-3}, with an average value of 30.6 µg m^{-3} in summer. Daily personal exposure to PM$_{2.5}$ varied in a range of 9.1-86.6 µg m^{-3} with an average value of 39.6 µg m^{-3} in winter. The intra-individual variability for each subject expressed with the coefficient of variance (CV%), were 62.0% and 48.9% in summer and winter respectively. Higher exposure levels were observed in winter among the subjects, and significant seasonal difference (*p*

= 0.003) was reported throughout these two sampling periods in Schedule A.

Table 3-3. Measured Personal $PM_{2.5}$ Exposure During Summer and Winter in Hong Kong

	Summer	Winter	Total
Mean[a]	30.6	39.6	34.9
SD[b]	19.0	19.4	19.6
Median	27.8	36.1	31.7
Minimum	3.5	9.1	3.5
Maximum	110.9	86.6	110.9
Q1	15.4	24.9	18.6
Q3	41.4	51.2	45.9
IQR[#]	25.9	26.3	27.3
CV (%)[c]	62.0	48.9	56.2
p-value[d]		0.003	
N[e]	87	80	167

Notes: [a]The unit of personal $PM_{2.5}$ mass concentration is µg m^{-3}; [b]SD refers to standard deviation; [c]CV (%) refers to the coefficient of variance; [d]The level of significance was taken as $p < 0.05$; [e]N refers to number of samples; [#]IQR refers to the interquartile range.
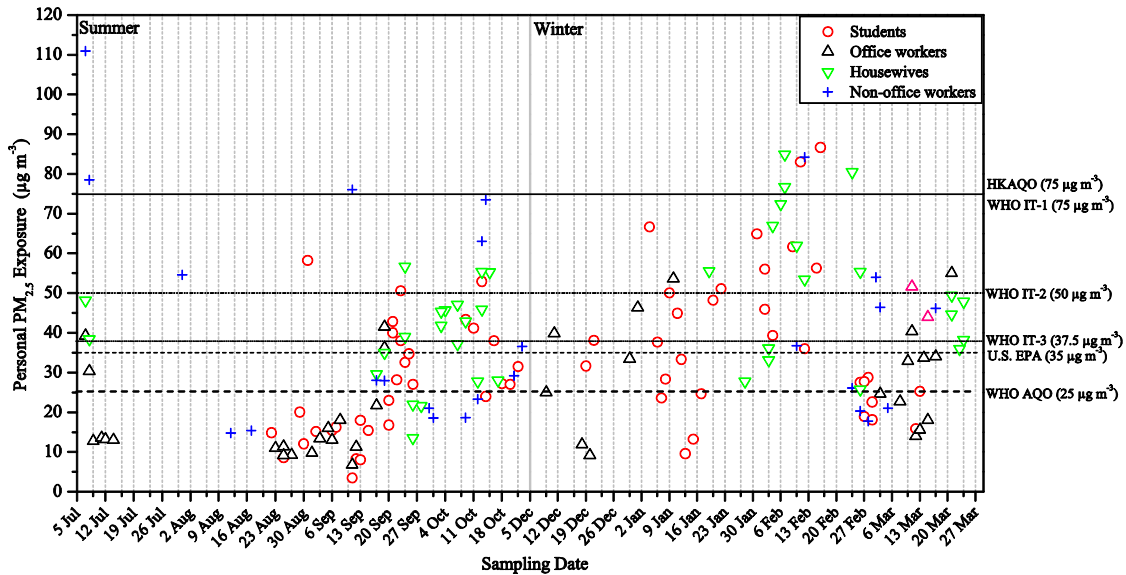
Figure 3-1. Personal $PM_{2.5}$ Mass Concentration During Summer and Winter Campaign Compared with WHO and U.S. EPA $PM_{2.5}$ 24-hr Ambient Air Quality Guideline

The $PM_{2.5}$ standards recommended by the "Air quality Guidelines" (issued by the World Health Organization (WHO) in 2005) are 25 µg m^{-3} within the 24-hr period and 10 µg m^{-3} for annual mean concentrations. The Hong Kong government announced newly proposed Air Quality Objectives (HKAQO) in January 2014, and the standards for $PM_{2.5}$ are an annual average of 35 µg m^{-3} and a 24-hr average of 75 µg m^{-3}. Besides the

guideline values, three interim targets (IT) are shown in Figure 3-1. 24-hr mean $PM_{2.5}$ concentrations of 75 μg m^{-3} was selected as the IT-1 level. The IT-2 interim levels of protection was set at 50 μg m^{-3}, and the recommended IT-3 level was 37.5 μg m^{-3}. For daily personal $PM_{2.5}$ mass concentrations, the median values were approximately half of the HKAQO 24-hr mean air quality guideline of 75 μg m^{-3} during the entire study period (Table 3-3 and Figure 3-1). The 24-hr average ambient $PM_{2.5}$ standard suggested by the World Health Organization (WHO) air quality guideline of 25 μg m^{-3} were exceeded in 52.8 % and 74.4% of all sampling day during summer and winter, respectively. In addition, the United States Environmental Protection Agency's (U.S. EPA) National Ambient Air Quality Standard (NAAQS) for 24-hr mean ambient $PM_{2.5}$ of 35 μg m^{-3} were exceeded in 37.1% of 87 personal exposures during summer and 52.4% of the 80 personal measurements in winter.

87 and 80 (valid samples) personal samples were obtained during summer and winter campaign, respectively, and personal $PM_{2.5}$ exposures among the general adult population in different gender, age, sampling day (workday and weekend) and residence areas in Hong Kong were investigated. As shown in Table 3-4, during the study period, comparable average personal $PM_{2.5}$ exposure levels were found for male (32.7 ± 20.1 μg m^{-3}) and female (37.6 ± 18.8 μg m^{-3}) subjects during the entire study period and no significant gender difference ($p = 0.110$) was reported. In general, higher exposure levels were found in a workday than on weekends during the study period, while no significant workday/weekend difference ($p = 0.087$) was observed. A summary of the activities of subjects in weekday and weekend during summer and winter is shown in Table S1 and Table S2. According to the results, the subjects spent an average of 63.5%-67.2% and 80.3 -87.3% of each day in their apartment during weekday and weekend, respectively. The time the subjects spent on transportation ranged from 5.4% to 6.1% and from 1.1% to 5.3% during the weekday the weekend, respectively. Higher $PM_{2.5}$ exposure levels in workdays may be due to more time spent on transportation and less time spent at home. Personal $PM_{2.5}$ average concentrations measured within the subjects' age 20-30 were the lowest exposure (27.6 ± 19.0 μg m^{-3}) ($p < 0.001$) out of all the subjects. Average personal $PM_{2.5}$ exposure levels for subjects residing in different districts varied in a range of 32.4-41.0 μg m^{-3} during the two sampling periods, and no significant spatial difference ($p = 0.206$) was found.

Table 3-4. Statistics for PM$_{2.5}$ Personal Exposure Measurement in Hong Kong

		Mean[a]	SD[b]	Median	Minimum	Maximum	p-value[c]	CV[d]	N[e]
Gender	Male	32.7	20.1	27.9	8.3	110.9	0.110	61.4%	92
	Female	37.6	18.8	38.3	3.5	83.0		50.1%	75
Age	< 20	39.8	17.9	38.0	15.5	86.6	< 0.001	45.0%	39
	20-30	27.6	19.0	23.3	3.5	110.9		69.0%	62
	30-50	34.2	18.1	28.1	12.8	73.4	< 0.001	52.9%	35
	>50	44.2	19.6	42.9	18.6	84.9	< 0.001	44.5%	31
Days	Workday	36.7	20.3	33.5	3.5	110.9	0.087	55.5%	111
	Weekend	31.4	17.8	27.6	8.1	84.9		56.6%	56
District (Home)	New Territories	32.4	19.0	28.3	8.6	84.9		58.6%	79
	Kowloon	36.2	18.8	34.7	3.5	86.6	0.206	51.9%	71
	Hong Kong Island	41.0	24.8	36.2	14.8	110.9		60.4%	17

Notes: [a]The unit of personal PM$_{2.5}$ mass concentration is µg m^{-3}; [b]SD refers to standard deviation; [c]The level of significance for the mass difference was taken as $p < 0.05$; [d]CV (%) refers to the coefficient of variance; [e]N refers to number of valid data.

As illustrated in Figure 3-2, significant difference ($p < 0.001$) was observed between different study groups during the entire sampling period, and significant higher exposure values were found for housewives and non-office workers compared with office workers. The average personal exposure to PM$_{2.5}$ for different study groups ranked in the descending order of housewives (44.4 ± 17.1 µg m^{-3}) ≈ non-office workers (40.3 ± 26.0 µg m^{-3}) > students (33.9 ± 18.2 µg m^{-3}) > officer workers (25.6 ± 14.6 µg m^{-3}). The subjects' activities were further examined with respect to their occupation characteristic in Table S3 and S4. The differences in PM$_{2.5}$ exposures between office workers and non-office workers reflected that non-office workers were used to spending more time on transportation or in outdoor places than office workers. The time the housewives spent inside their apartment, especially in the kitchen, equals 6.4%-9.8% of an average 24-hr day. Aerosol generated during the cooking process, typically representing a significant PM$_{2.5}$ source, might have a significant influence on individual PM$_{2.5}$ exposure levels.
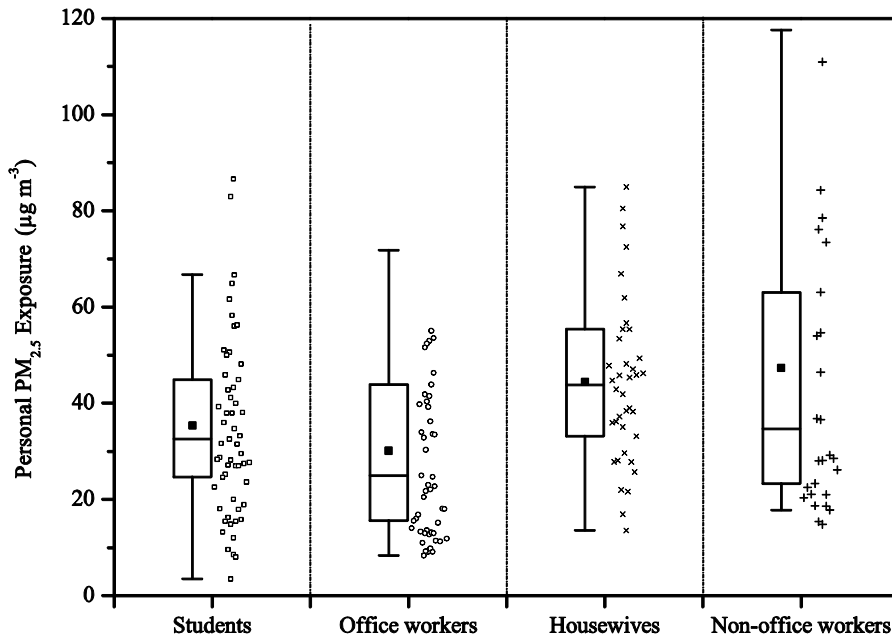
Figure 3-2. Variations of Personal Exposure to PM$_{2.5}$ for Students, Office Workers, Housewives and Non-Office Workers

Exposures can vary across locations and within persons over time (due to differences in activities and environmental factors). Relatively higher exposure levels were found in winter than that in summer, and significant seasonal variations were reported for male ($p = 0.029$) and female ($p = 0.018$) subjects (Figure 3-3). Comparable higher personal PM$_{2.5}$ exposure levels were reported in winter than that in summer during the workday and weekend, respectively. Personal PM$_{2.5}$ average concentrations measured within the subjects of different age showed higher average exposure levels in winter (range: 30.6-53.5 µg m^{-3}) than in summer (range: 24.4-37.5 µg m^{-3}). In general, higher exposure levels were found in winter for subjects residing in different districts of Hong Kong as shown in Figure 3-3.

When comparing the seasonal variations of personal PM$_{2.5}$ exposure levels for the occupation of different subjects, the lowest concentrations were observed among office workers, both in summer (20.6 ± 13.0 µg m^{-3}) and winter (32.4 ± 14.3 µg m^{-3}). According to the results of previous studies in Hong Kong and the measurements in the following

section, significant higher ambient PM$_{2.5}$ concentrations were reported in winter than in summer. Despite subjects spent a majority of their time in the indoor environment during the 24-hr day, personal exposure level is still influenced by the ambient air quality. During these two monitoring periods, higher exposure levels were observed in winter than that in summer, and significant seasonal differences were observed among office workers ($p = 0.006$), students ($p = 0.017$) and housewives ($p = 0.025$) in this study.
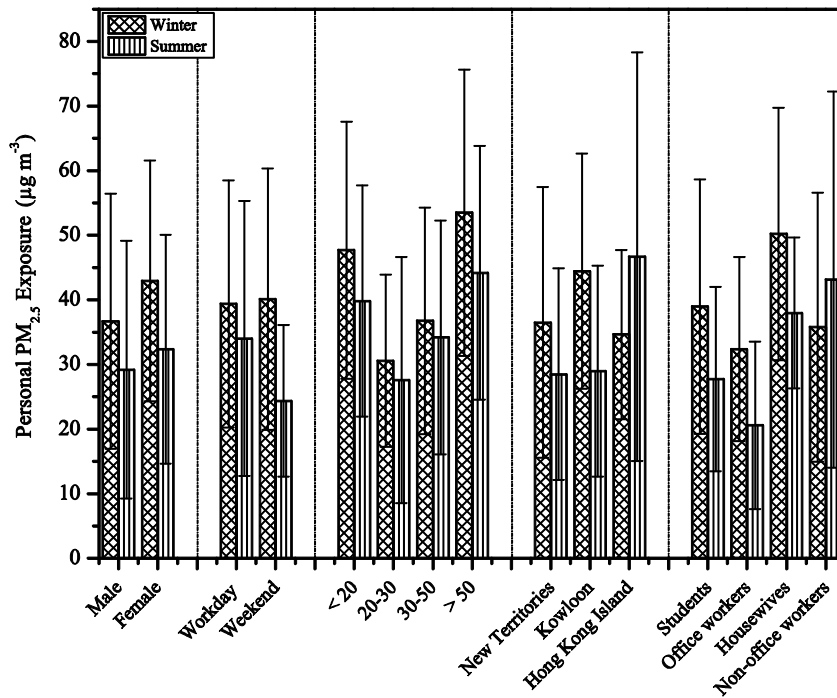
Figure 3-3. Seasonal Variations of Personal Exposure to PM$_{2.5}$ in Hong Kong
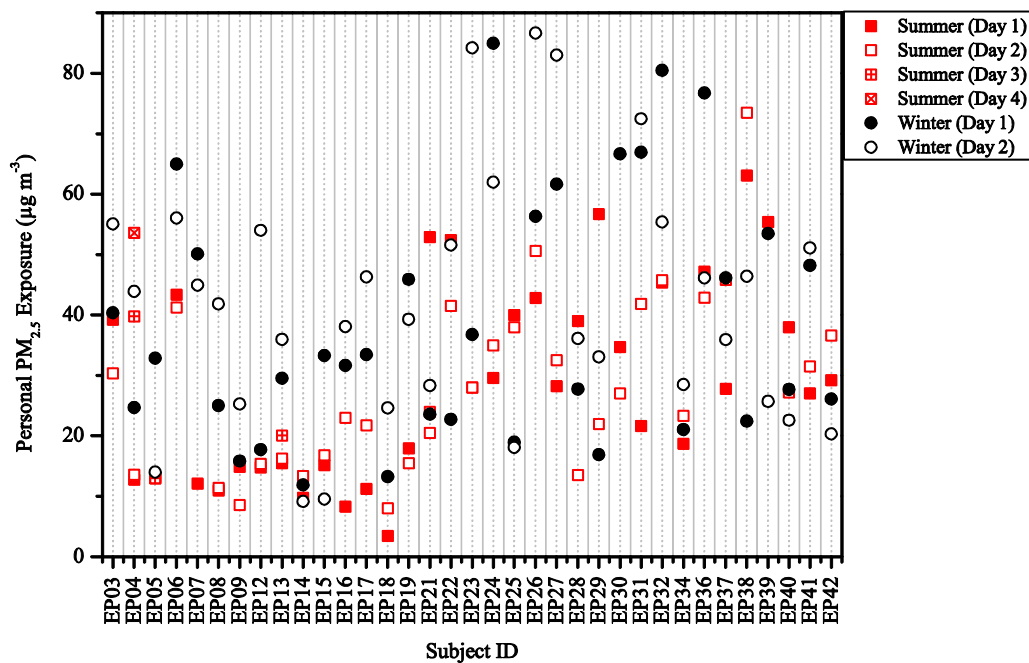
Figure 3-4. Individual Personal PM$_{2.5}$ Exposure

Scatter plots of individual PM$_{2.5}$ exposures from the sampling days of personal monitoring by subjects participated both in summer and winter campaign (N = 35) are shown in Figure 3-4. This figure also relates to the within-subject factor (mentioned in Section 2.2.4). The variability (CV%) of personal exposure for subjects (EP04, EP05, EP13, and EP21) between two sampling days ranged from 0.9% to 67.3%. Subject EP38 and EP42 were found to have higher exposures in summer than that in winter during two sampling days (Figure 3-4), which are reflected in their observed variability plots within the graph. Ambient PM$_{2.5}$ displayed significant seasonal variations with higher values in winter and lower values in summer (Section 3.2) while slightly higher personal PM$_{2.5}$ were reported in summer than that measured in winter for non-office workers. Based on activity data in Table S3 and Table S4, the differences in PM$_{2.5}$ exposures for non-office workers in summer and winter reflected that subjects were used to spending more time on transportation or in other indoor places (e.g., restaurant) in summer than in winter. However, only 10 measurements from non-office worker subjects (e.g., EP12, EP23, EP34, EP38, EP42) was obtained in winter during this study, and the results may not

explain the variation in exposure that from inter-individual (between subject) factors (e.g., factors affect all of the subjects). Thus, further study relates to the occupation of different subjects should be performed in future.

- Regression analysis of pooled data

A major objective of the Schedule A was to investigate the seasonal and spatial variation of personal exposure to $PM_{2.5}$ in Hong Kong thus ambient measurements were not conducted during the personal sampling period. Therefore, daily average ambient $PM_{2.5}$ concentrations during the study period in Schedule A were calculated from the Air Quality Monitoring data based on the daily ambient concentrations (14 ambient monitoring sites including 11 general stations and 3 roadside stations) archived by the Hong Kong Environmental Protection Department (HKEPD, http://epic.epd.gov.hk/EPICDI/air/download/). The Pearson correlation r between personal $PM_{2.5}$ exposure and ambient concentrations were 0.66 both in summer and winter, respectively, in Hong Kong.
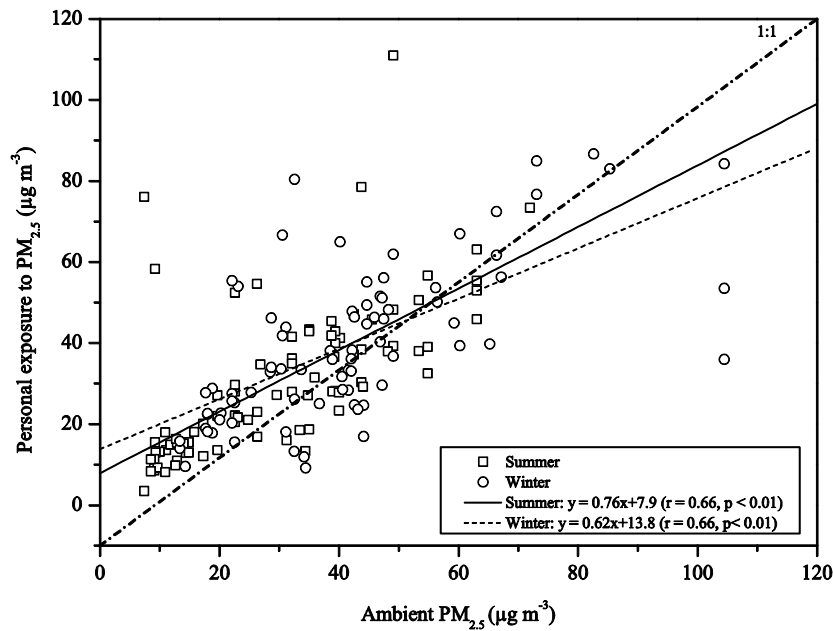
Figure 3-5. The relationship between the individual personal exposure to $PM_{2.5}$ and the ambient concentrations of $PM_{2.5}$ during summer and winter in Hong Kong.

- Chemical Species in Personal $PM_{2.5}$

    The average concentrations of chemical species including organic carbon, elemental carbon, water-soluble ions and elements in personal $PM_{2.5}$ during summer and winter in Hong Kong were presented in Table 3-5. Seasonal variations (W/S ratio) were shown for most components in personal $PM_{2.5}$, and higher concentrations were observed in winter.

Table 3-5. Average Concentrations of Chemical Species in Personal $PM_{2.5}$ Samples

Species[a]	Summer (S)			Winter (W)			Total			W/S ratio[d]
	Mean	SD[b]	N[c]	Mean	SD	N	Mean	SD	N	Mean $\pm$ SD
OC	**7.43**	5.05	87	**8.04**	4.65	80	7.72	4.86	167	1.3 $\pm$ 0.7
EC	**2.08**	1.12	87	**2.28**	1.03	80	2.18	1.08	167	1.4 $\pm$ 0.8
Na^+	0.56	0.29	83	0.58	0.27	78	0.57	0.28	161	1.3 $\pm$ 0.8
NH_4^+	**3.11**	2.35	82	**5.04**	2.65	80	4.06	2.67	162	3.2 $\pm$ 4.6
K^+	0.26	0.17	82	0.36	0.24	80	0.31	0.22	162	2.1 $\pm$ 1.7
Mg^{2+}	0.05	0.03	49	0.08	0.04	80	0.07	0.04	129	2.3 $\pm$ 1.6
Ca^{2+}	0.34	0.83	86	0.22	0.14	77	0.28	0.61	163	1.5 $\pm$ 1.6
Cl^-	0.22	0.28	87	0.61	0.66	78	0.40	0.54	165	4.9 $\pm$ 6.0
NO_3^-	**1.07**	1.23	87	**4.94**	4.72	80	2.93	3.89	167	7.8 $\pm$ 7.9
SO_4^{2-}	**7.92**	5.63	87	**11.28**	5.58	80	9.53	5.84	167	2.3 $\pm$ 2.1
Na	3.100	2.167	81	3.765	2.114	79	3.428	2.160	160	2.7 $\pm$ 5.6
Mg	0.140	0.095	56	0.157	0.104	55	0.149	0.100	111	1.9 $\pm$ 2.5
Al	0.114	0.124	80	0.158	0.098	73	0.135	0.114	153	3.5 $\pm$ 4.2
Si	0.253	0.497	82	0.252	0.196	79	0.252	0.380	161	5.3 $\pm$ 15.4
S	2.416	1.688	86	3.041	1.305	80	2.717	1.543	166	3.2 $\pm$ 5.5
Cl	0.083	0.248	82	0.286	0.323	79	0.182	0.304	161	10.8 $\pm$ 15.5
K	0.246	0.211	86	0.407	0.259	80	0.323	0.248	166	4.0 $\pm$ 5.5
Ca	0.389	1.492	85	0.169	0.116	80	0.283	1.077	165	1.9 $\pm$ 2.5
Ti	0.014	0.020	84	0.015	0.013	77	0.014	0.017	161	2.7 $\pm$ 4.2
V	0.015	0.015	85	0.014	0.016	76	0.014	0.016	161	2.2 $\pm$ 4.1
Cr	0.004	0.002	27	0.003	0.002	59	0.003	0.002	86	1.2 $\pm$ 0.8
Mn	0.014	0.016	77	0.014	0.008	79	0.014	0.013	156	2.7 $\pm$ 4.1
Fe	0.324	0.403	86	0.268	0.201	80	0.297	0.322	166	6.1 $\pm$ 24.3
Ni	0.005	0.004	69	0.005	0.004	77	0.005	0.004	146	2.0 $\pm$ 3.1
Cu	0.019	0.017	79	0.028	0.027	80	0.023	0.023	159	4.0 $\pm$ 9.5
Zn	0.142	0.287	86	0.125	0.097	80	0.134	0.217	166	5.6 $\pm$ 12.4
As	0.002	0.001	50	0.004	0.002	55	0.003	0.002	105	1.7 $\pm$ 1.3
Br	0.009	0.006	82	0.019	0.014	80	0.014	0.012	162	4.3 $\pm$ 5.5
Pb	0.026	0.018	62	0.031	0.025	80	0.029	0.022	142	2.6 $\pm$ 3.9

Notes: [a]The unit of personal $PM_{2.5}$ mass concentration and chemical species is µg m^{-3}; [b]SD refers to standard deviation; [c]N refers to number of valid data and values below the MDL have been discarded; [d] W/S = Winter/Summer

OC and EC

The average OC and EC concentrations in personal $PM_{2.5}$ during summer were 7.43 ± 5.05 and 2.08 ± 1.12 µg m^{-3}, respectively (Table 3-5). Carbonaceous compositions showed the similar seasonal pattern as personal $PM_{2.5}$ mass concentrations, and relatively higher average values were found for OC (8.04 ± 4.65 µg m^{-3}), and EC (2.28 ± 1.03 µg m^{-3}) in personal $PM_{2.5}$, respectively, in winter. While no significant seasonal variations were found for OC ($p = 0.422$) and EC ($p = 0.250$). As reported in Table 3-6, OC and EC accounted for an average 27.7% and 7.7% of personal $PM_{2.5}$ concentrations in summer. Personal $PM_{2.5}$ consisted of 21.8% OC and 6.3% EC in winter.

Table 3-6. Statistical Description of the Concentrations of OC and EC in Personal $PM_{2.5}$

	Summer	Winter	Total
OC/EC (no unit)	3.9 ± 1.6 [a]	3.7 ± 2.1	3.8 ± 1.8
TC/Personal $PM_{2.5}$ (%)[b]	35.3 ± 12.9	28.1 ± 14.5	31.9 ± 14.1
OC/Personal $PM_{2.5}$ (%)	27.7 ± 11.4	21.8 ± 13.3	24.9 ± 12.6
EC/Personal $PM_{2.5}$ (%)	7.7 ± 2.9	6.3 ± 2.8	7.0 ± 2.9
N[c]	87	80	167

Notes: [a]Values represent average $\pm$ standard deviation; [b]Percentage of carbonaceous aerosol concentration to personal $PM_{2.5}$ mass concentration; [c]N refers to number of valid data.

Typically, EC is emitted from combustion sources, while OC can be emitted from both primary sources and generated from chemical reactions among gaseous organic carbon species. OC/EC ratio has been used to identify the presence of secondary organic aerosol when the OC/EC ratio exceeding 2. Moderate to high correlations between OC and EC in personal $PM_{2.5}$ for different subjects were observed in summer (R^2: 0.20-0.66) and winter (R^2: 0.34-0.62). Average personal OC/EC ratios were 3.9 and 3.7 during summer and winter, respectively. The OC/EC ratio was further examined with respect to their occupation characteristics. As shown in Table 3-7, average OC/EC ratios varied from 3.3 to 4.2 for personal measurements. Ambient OC/EC ratios ranged from 1.2 (roadside: Mong Kok) to 3.0 (sub-urban: Clear Water Bay) during the same period when personal samples were collected. It seems that there is a significant secondary OC contribute in the personal samples. In contrast, lower OC/EC ratios were observed in the indoor environment (2.2-3.1) and in the outdoors (1.7-2.4) in Hong Kong (Ho et al., 2004).

Table 3-7. Seasonal Variations of OC and EC in Personal $PM_{2.5}$ for Different Study Groups in Hong Kong

		OC^b	EC	OC/EC (no unit)	OC/Personal $PM_{2.5}$ (%)^c	EC/Personal $PM_{2.5}$ (%)	N^d
Summer	Students	6.7 ± 2.3	1.8 ± 0.8	4.3	30.0	7.2	29
	Office workers	**5.4 ± 2.3**	**1.6 ± 1.0**	3.9	30.3	8.7	23
	Housewives	8.0 ± 2.8	2.4 ± 1.0	3.5	21.7	6.6	19
	Non-office workers	10.6 ± 9.4	**2.8 ± 1.4**	3.7	26.5	8.0	16
Winter	Students	8.9 ± 5.1	2.2 ± 0.8	4.0	23.0	6.3	28
	Office workers	**5.7 ± 2.6**	**1.6 ± 0.7**	4.0	21.9	5.3	23
	Housewives	9.8 ± 5.4	2.7 ± 1.1	3.7	19.4	5.3	19
	Non-office workers	7.6 ± 3.9	**3.2 ± 1.1**	2.5	23.4	10.3	10
Total	Students	7.7 ± 4.1	1.9 ± 0.8	4.2	26.6	6.8	57
	Office workers	**5.6 ± 2.5**	**1.6 ± 0.9**	3.9	26.0	6.9	46
	Housewives	8.9 ± 4.4	2.5 ± 1.0	3.6	20.5	6.0	38
	Non-office workers	**9.5 ± 7.9**	**2.9 ± 1.3**	3.3	25.4	8.8	26

Notes: [a]The unit of OC and EC in personal $PM_{2.5}$ is µg m^{-3}; [b]Values represent average $\pm$ standard deviation; [c]Percentage of carbonaceous aerosol concentration to personal $PM_{2.5}$ mass concentration; [d]N refers to number of valid data.

As shown in Table 3-7, average OC and EC concentrations in personal $PM_{2.5}$ for different study groups ranked in the ascending order of office workers < students < housewives < non-office workers. During these two monitoring periods, significant difference was observed for office workers and non-office workers, and average EC concentrations in personal $PM_{2.5}$ for non-office workers were about two times higher than office workers. It should be noted that according to the personal activity diaries, non-office worker subjects (e.g., van drivers who spent 180-780 min/day on transportation) had the highest EC exposure levels while office workers (> 8h in the office on a workday) had the lowest EC exposures. Results in Table S3 and S4 also show the non-office workers, on average, spent 15.8%-17.5% of their time in a van and 6.2-15.9% outdoor. In contrast, office workers spent only 0.7-1.7% of their time on the bus/minibus and 2.0-2.4% outdoor. Personal $PM_{2.5}$ consisted of 6.9% EC for office worker and 8.8% EC for non-office worker subjects. The highest percentage of EC in personal $PM_{2.5}$ was observed for non-office workers in winter (10.3%), which was 1.9 times higher than those measured for office workers (Table 3-7).

Inorganic Ions

As shown in Table 3-5, sulfate (SO_4^{2-}) was the most abundant chemical component in personal $PM_{2.5}$. Average personal sulfate accounted for 27.3% of the personal $PM_{2.5}$ mass. On average, SO_4^{2-} concentration in winter was significant 1.4 times higher than that in summer in personal $PM_{2.5}$ samples. Similar seasonal variation was also reported for other inorganic ions. The most abundant ionic species in personal $PM_{2.5}$ followed the order of $SO_4^{2-} > NH_4^+ > NO_3^- > Na^+ > K^+ > Mg^{2+} > Ca^{2+}$. The secondary inorganic aerosols ($\sum$SIA, the sum of SO_4^{2-}, NH_4^+, and NO_3^-) and their average values in personal $PM_{2.5}$ were 11.92 ± 8.66 and 21.27 ± 12.32 µg m^{-3}, respectively, in summer and winter. The secondary inorganic aerosols in personal $PM_{2.5}$ for different study groups followed the order of $SO_4^{2-} > NH_4^+ > NO_3^-$. In line with the $PM_{2.5}$ exposure levels, the average SIA in personal exposure $PM_{2.5}$ for different study groups ranked in the descending order of housewives > non-office workers > students > officer workers. Secondary inorganic ions accounted for 38.9% of personal $PM_{2.5}$ in Hong Kong during summer. While SIA was the largest contributor in winter, accounted for 53.0% of personal $PM_{2.5}$. High percentage of water-soluble ions in winter and high percentage of carbonaceous species in summer were observed at urban sites in Hong Kong (Ho et al., 2006b). The seasonal difference of personal $PM_{2.5}$ was most due to carbonaceous aerosols and secondary inorganic ions. Major composition and source analysis indicated that the increased $PM_{2.5}$ mass concentrations in winter may be caused by the long-range transportation of air pollution from mainland China (Ho et al., 2006b). For sulfate, there is limited evidence of indoor or personal sources, and personal exposures were a fraction of the ambient levels (Noullett et al., 2006). Similar situation was observed for nitrate and ammonium. The higher SIA concentration in personal $PM_{2.5}$ may due to the regional sources contribution in winter.

Elements

Generally speaking, crustal elements (e.g., Na, Mg, Al, Si, Ca, Ti, Fe) have relatively high proportions of the target elements (Table 3-5). Significantly higher concentrations were recorded for sulfur in personal $PM_{2.5}$ during summer (2.4 µg m^{-3}) and winter (3.0 µg m^{-3}). Crustal elements accounted for over 90.7% (range: 89.3%-92.3%) of the total elements during the whole study period. Trace elements such as V, Cr, Mn, Ni,

Cu, Zn, As, and Pb had lower concentrations and accounted for less than 5% (range: 2.5%-3.1%) of the total elements. Concentrations of Zn and Pb measured in personal $PM_{2.5}$ samples were higher when compared to other trace elements (e.g., As, Br, Cr).

Generally speaking, the concentrations of elements determined in this study for different study groups followed the order of housewives > non-office workers > students > officer workers in summer. The similar descending order was observed in winter. When comparing the seasonal variations of elements in personal $PM_{2.5}$ for the occupation of different subjects, the lowest concentrations were observed among office workers both in summer and winter.

Significant higher values of trace elements (Zn and Mn) for non-office workers (e.g., van drivers) were reported in Table 3-8 during the study period, suggesting a tire wear origin. Higher crustal materials (e.g., Mg, Al, Si, Ca, Fe) were also observed for non-office workers and housewives compare to other subjects in summer and winter, respectively. The trace metals Ni and V, suggesting an oil combustion sources for housewives.

Table 3-8. Average concentrations and standard deviations of ions and major elements in Personal PM2.5 for Different Study Groups in Hong Kong.

		Mean ± SD				Normalization (%)			
		Students	Office workers	housewives	Non-office workers	Students	Office workers	Housewives	Non-office worker
	Summer								
Ions (μg m^{-3})	Na$^+$	0.5 ± 0.4	0.5 ± 0.3	0.7 ± 0.2	0.5 ± 0.2	-3.5%	-9.6%	21.6%	-5.4%
	NH$_4^+$	2.7 ± 2.0	1.6 ± 1.5	4.9 ± 2.2	2.8 ± 2.9	-7.3%	-44.6%	68.2%	-3.7%
	K$^+$	0.2 ± 0.1	0.1 ± 0.1	0.3 ± 0.1	0.3 ± 0.2	-7.0%	-44.5%	40.9%	28.1%
	Mg^{2+}	0.0 ± 0.0	0.0 ± 0.0	0.0 ± 0.0	0.0 ± 0.0	-27.8%	-47.8%	55.8%	52.8%
	Ca^{2+}	0.2 ± 0.1	0.2 ± 0.2	0.3 ± 0.1	1.0 ± 1.8	-49.1%	-48.6%	-23.7%	187.2%
	Cl$^-$	0.2 ± 0.3	0.3 ± 0.4	0.1 ± 0.1	0.2 ± 0.2	12.4%	25.4%	-35.3%	-16.9%
	NO$_3^-$	0.8 ± 0.8	0.9 ± 1.4	1.5 ± 1.4	1.2±1.4	-23.6%	-14.1%	43.1%	11.8%
	SO$_4^{2-}$	7.4 ± 5.0	4.7 ± 3.4	12.4 ± 5.2	8.2±6.6	-7.2%	-40.4%	56.4%	4.1%
Elements (ng m^{-3})	Na (μg m^{-3})	2.7 ± 1.7	1.9 ± 1.6	5.2 ± 2.0	3.0 ± 2.2	-13.7%	-38.1%	67.4%	-4.1%
	Mg	136.7 ± 82.5	165.7 ± 109.2	156.9 ± 92.5	76.8 ± 54.3	-3.3%	17.2%	11.1%	-45.6%
	Al	92.8 ± 70.5	58.0 ± 70.8	157.5 ± 75.5	179.5 ± 216.2	-18.2%	-48.9%	38.9%	58.2%
	Si	141.4 ± 124.7	119.8 ± 213.5	281.1 ± 188.6	567.7 ± 1,021.5	-42.7%	-51.4%	14.0%	130.1%
	S (μg m^{-3})	2.1 ± 1.4	1.6 ± 1.4	3.9 ± 1.4	2.6 ± 12.0	-13.5%	-34.2%	60.6%	6.1%
	Cl	118.8 ± 421.6	66.1± 141.7	41.3 ± 19.5	88.7 ± 89.5	47.7%	-17.8%	-48.7%	10.2%
	K	199.3 ± 149.5	141.7 ± 138.6	332.4 ± 158.3	386.1 ± 322.9	-18.4%	-42.0%	36.1%	58.1%
	Ca	134.2 ± 101.8	150.4 ± 221.1	178.1 ± 111.9	1,396.8 ± 3,319.7	-64.3%	-60.0%	-52.6%	271.3%
	Ti	9.8 ± 7.4	9.1 ± 10.9	14.2 ± 7.1	27.2 ± 40.0	-28.7%	-34.0%	3.5%	97.8%
	V	12.9 ± 10.7	14.7 ± 15.6	17.7 ± 16.7	13.7 ± 18.8	-12.3%	0.4%	<u>20.6%</u>	-6.3%
	Cr	4.8 ± 2.9	3.4 ± 2.6	2.7 ± 1.6	3.0 ± 1.9	33.9%	-5.4%	-24.2%	-16.6%
	Mn	11.2 ± 7.4	8.9 ± 7.9	13.5 ± 6.6	21.7 ± 31.4	-14.5%	-32.2%	3.3%	<u>65.3%</u>
	Fe	341.3 ± 376.6	203.5 ± 228.5	278.0 ± 280.1	532.5 ± 650.7	7.2%	-36.1%	-12.7%	67.3%
	Ni	3.8 ± 2.6	4.5 ± 4.2	5.2 ± 4.6	5.0 ± 6.1	-16.3%	-1.6%	<u>14.0%</u>	9.3%
	Cu	17.8 ± 21.3	19.7 ± 19.8	17.6 ± 13.8	16.7 ± 11.0	-2.0%	8.8%	-2.6%	-7.7%
	Zn	117.0 ± 154.4	89.5 ± 145.6	119.9 ± 60.4	284.8 ± 599.0	-15.8%	-35.6%	-13.7%	<u>105.0%</u>
	As	2.5 ± 1.1	1.9 ± 0.9	2.6 ± 1.1	2.8 ± 1.3	-0.3%	-23.7%	6.8%	12.4%
	Br	7.7 ± 5.5	7.0 ± 6.1	12.7 ± 4.9	9.6 ± 6.6	-13.8%	-22.1%	42.4%	7.7%
	Pb	25.5 ± 17.2	17.8 ± 14.9	32.6 ± 18.8	24.3 ± 18.0	-0.1%	-30.4%	27.4%	-4.9%
	Winter								
Ions (μg m^{-3})	Na$^+$	0.6 ± 0.3	0.5 ± 0.3	0.6 ± 0.2	0.5 ± 0.3	6.8%	-14.4%	11.3%	-7.3%
	NH$_4^+$	4.8 ± 2.5	4.2 ± 2.0	6.9 ± 2.6	4.0 ± 2.8	6.2%	-8.6%	4.6%	-6.2%
	K$^+$	0.4 ± 0.3	0.2 ± 0.1	0.4 ± 0.2	0.3 ± 0.3	-3.9%	-17.3%	37.5%	-20.7%

	Mg^{2+}	0.1 ± 0.0	0.1 ± 0.0	0.1 ± 0.0	0.1 ± 0.0	15.3%	-32.2%	19.5%	-5.8%
	Ca^{2+}	0.2 ± 0.1	0.2 ± 0.1	0.2 ± 0.1	0.3 ± 0.2	-3.3%	-9.4%	8.3%	15.2%
	Cl^-	0.7 ± 0.9	0.4 ± 0.3	0.7 ± 0.6	0.4 ± 0.5	-11.6%	-12.9%	14.4%	34.8%
	NO_3^-	4.9 ± 5.1	3.5 ± 2.5	7.4 ± 5.6	3.8 ± 4.5	17.9%	-26.1%	18.7%	-25.6%
	SO_4^{2-}	11.3 ± 5.8	9.0 ± 4.2	15.0 ± 5.1	9.4 ± 5.5	-0.7%	-30.0%	49.3%	-22.7%
Elements (ng m⁻³)	Na (μg m⁻³)	3.6 ± 2.1	3.1 ± 1.8	5.0 ± 2.1	3.6 ± 2.3	0.2%	-20.6%	33.3%	-16.3%
	Mg	158.0 ± 121.3	131.3 ± 68.9	184.5 ± 101.6	122.9 ± 106.6	5.7%	-24.3%	24.9%	-7.4%
	Al	149.2 ± 89.1	150.9 ± 102.3	197.5 ± 118.3	145.5 ± 76.6	-7.9%	-6.9%	21.9%	-10.2%
	Si	265.4 ± 206.9	263.6 ± 182.6	299.5 ± 262.2	190.0 ± 145.0	0.5%	-0.2%	13.4%	-28.1%
	S (μg m⁻³)	2.9 ± 1.3	2.7 ± 1.2	3.9 ± 1.0	2.6 ± 1.5	-4.4%	-12.9%	28.2%	-16.7%
	Cl	300.5 ± 380.6	241.5 ± 175.9	331.9 ± 318.6	266.9 ± 387.7	3.5%	-16.8%	14.4%	-8.0%
	K	474.8 ± 329.7	297.6 ± 150.2	458.0 ± 193.0	382.9 ± 272.5	14.1%	-28.5%	10.0%	-8.0%
	Ca	180.8 ± 213.7	179.4 ± 157.0	173.0 ± 86.3	221.9 ± 138.2	-1.5%	-2.3%	-5.7%	20.9%
	Ti	13.9 ± 8.8	17.8 ± 23.2	15.9 ± 8.1	15.4 ± 8.5	-10.0%	14.9%	2.9%	-0.5%
	V	12.2 ± 14.6	11.8 ± 14.2	21.0 ± 23.8	9.3 ± 4.5	-12.0%	-14.8%	<u>51.1%</u>	-33.0%
	Cr	3.0 ± 1.9	3.7 ± 2.7	2.5 ± 1.5	3.7 ± 2.4	-5.0%	17.7%	-19.8%	17.6%
	Mn	13.3 ± 6.7	13.4 ± 7.5	14.9 ± 6.7	16.2 ± 15.0	-5.4%	-4.8%	6.1%	<u>15.2%</u>
	Fe	269.0 ± 226.9	304.4 ± 289.1	230.2 ± 118.7	370.2 ± 189.6	-4.1%	8.5%	-17.9%	32.0%
	Ni	3.9 ± 3.8	4.6 ± 4.0	6.4 ± 6.1	3.3 ± 1.1	-16.1%	1.1%	<u>40.5%</u>	-27.3%
	Cu	31.1 ± 29.6	23.2 ± 18.9	34.2 ± 34.1	18.6 ± 11.4	9.7%	-18.4%	20.2%	-34.5%
	Zn	129.9 ± 87.4	134.2 ± 98.9	122.1 ± 76.8	132.8 ± 173.6	0.4%	3.7%	-5.7%	<u>2.7%</u>
	As	4.3 ± 2.3	2.7 ± 1.5	4.1 ± 2.0	2.5 ± 2.8	14.9%	-27.6%	11.6%	-32.6%
	Br	23.5 ± 16.4	15.1 ± 9.9	21.8 ± 13.4	15.0 ± 13.9	17.7%	-24.8%	9.1%	-25.3%
	Pb	35.3 ± 25.2	27.5 ± 22.6	36.6 ± 27.2	22.1 ± 25.6	9.9%	-14.3%	14.0%	-31.2%

* Normalization = (average- total average)/total average × 100%
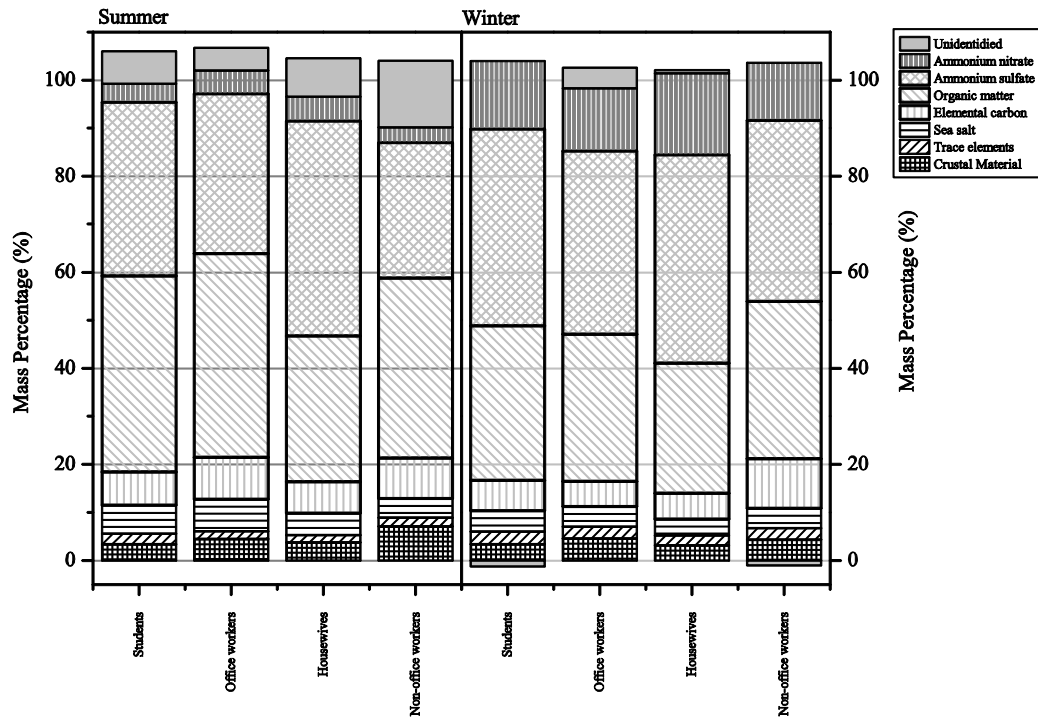
3-16

Figure 3-6. Personal PM$_{2.5}$ Material Balance Among Different Subjects During Summer and Winter in Hong Kong. Negative Unidentified Mass Fraction Indicates A Greater Reconstructed Mass than Measured Gravimetric Mass

Average mass fractions (equation 2.1) of chemical species in personal PM$_{2.5}$ were shown, during summer and winter, in Figure 3-4. The largest fraction in personal PM$_{2.5}$ was carbonaceous materials (summer: 36.9%-51.1%; winter: 32.4%-43.0%). Organic matter contributions accounted for a third of personal PM$_{2.5}$ mass during summer (30.3-42.4%) and winter (27.1-32.7%). Secondary ammonium sulfate was the largest contributor in winter, accounting for more than a third (37.7-43.3%) of personal PM$_{2.5}$ mass. Most of the elements (i.e., crustal and trace elements) had higher mass percentages among non-office workers. Sea salt accounted for 3.4-6.7% of personal PM$_{2.5}$ mass.

3.2   Schedule B

3.2.1 _**Study Group**_

The study group consisted of 12 adults, 5 women and 7 men between 19 and 57 years of age and living in different districts of Hong Kong (Table 3-9). 3 Groups collected 24-hr personal PM2.5 samples following a schedule of 4 samples for every six days, and the sampling dates were in accordance with the HKEPD $PM_{2.5}$ sampling schedule. All subjects were all non-smokers and not featured any chronic diseases. Occupation of all the subjects was categorized into 4 groups (Table 3-8), students (3 subjects), office workers (3 subjects), housewives (3 subjects) and non-office workers (3 subjects).

Table 3-9. Description of the Subjects for the Personal Sampling of Schedule B

	One year sampling campaign
Sampling Date	April 25, 2014 – June 7, 2015
Study subjects (N)	12
Female	5
Male	7
Median age, years (range)	37 (19-57)
Occupation (N)	
Students	3
Office workers	3
Housewives	3
Non-office workers	3
Group (Collected samples, N)	248
Group 1	82
Group 2	84
Group 3	82

3.2.2 _**Activity Profiles**_

A summary of subjects' personal activity pattern was reported in Table 3-9. Time-activity diaries were completed every 30 min for each of the monitoring days. The monitoring parameters included time at home, workplace, outdoors and time spent on transportation (e.g., bus, minibus, metro). Moreover, daily activity patterns of each subject during the one-year sampling period were shown in Table S5-S6 (Appendix I). A total of 229 (92.3%) valid daily activity data was collected in Schedule B (Table 3-10).

According to the one-year long activity diaries, the subjects spent an average of 83.7% of each day in an indoor environment. The average daily time contribution of time spent at home was equal to 71.2% (average of 69.4% and 75.6% during weekday and weekend, respectively (Table S5)). As shown in Table 3-10, a considerable portion of time was spent outdoors (11.9%) with the rest of the daily activities divided between transportation (4.0%) and indoors at work (4.8%) for all subjects. Variation of the profile did occur with individuals spending an average of 54.3-96.3% of their time in the apartment, and a considerable portion of time (ranged from 2.6% to 32.4%) in the outdoors (Table S6).

Table 3-10. Subjects' Activity During the Personal Sampling Campaign (Schedule B)

Time-activity data from diaries (N = 229)	Average	Median
Time spent (%)		
Indoors, total	83.7	87.5
Indoors at home	71.2	70.8
Indoors at work (average)	4.8	0.0
Outdoors	11.9	7.0
Transportation	4.0	3.5

3.2.3 *Person PM$_{2.5}$ Exposure*

Ambient PM$_{2.5}$ concentrations were simultaneously measured from seven EPD monitoring stations at different areas in Hong Kong, and the personal PM$_{2.5}$ sampling date matched with the ambient PM$_{2.5}$ sampling schedule. Regression analysis determined correlations between ambient PM$_{2.5}$ measured at different monitoring sites and total personal PM$_{2.5}$. Their correlation coefficients were listed in Table 3-11.

Table 3-11. Summary of Correlation Coefficients of Ambient PM$_{2.5}$ and Personal PM$_{2.5}$

Monitoring Stations	Ambient PM$_{2.5}$							Personal PM$_{2.5}$
	MK	CW	WB	TC	TW	YL	KC	
Mong Kok (MK)	1	0.987**	0.957**	0.904**	0.934**	0.919**	0.937**	0.832**
Central/Western (CW)		1	0.956**	0.924**	0.951**	0.935**	0.947**	0.844**
Clear Water Bay (WB)			1	0.940**	0.925**	0.932**	0.916**	0.821**
Tung Chung (TC)				1	0.958**	0.985**	0.948**	0.838**
Tsuen Wan (TW)					1	0.969**	0.982**	0.833**
Yune Long (YL)						1	0.958**	0.852**
Kwai Chung (KC)							1	0.852**
Personal PM$_{2.5}$								1

**Correlation is significant at the 0.01 level (2-tailed)

Significant correlations (r_s range: 0.904-0.987) were observed between ambient PM$_{2.5}$ at the seven monitoring sites. Correlation coefficients between ambient PM$_{2.5}$ and total personal PM$_{2.5}$ ranged from 0.821 to 0.852 ($^{**}p < 0.01$). The descriptive statistics of the seasonal variability observed in personal PM$_{2.5}$ exposures and ambient concentrations were shown in Table 3-12.

Table 3-12. Seasonal Variations and Mass Concentration Differences Between Ambient and personal Exposure to PM$_{2.5}$ in Hong Kong

Personal (P)[a] Ambient (A)[b] P/A ratio (no unit)	Mean	SD[c]	Median	Minimum	Maximum	N[d]	Mass Difference *p-value*[e]
Spring	31.2	16.0	27.2	10.1	74.7	80	< 0.001
	23.3	11.5	22.0	5.4	58.2	172	
	1.5	0.6	1.3	0.5	3.5	146	
Summer	22.5	11.8	18.3	6.8	56.2	65	< 0.001
	14.1	5.6	12.9	6.5	35.0	138	
	1.8	0.8	1.6	0.5	4.5	110	
Autumn	34.6	12.0	36.5	11.1	62.2	61	0.254
	33.1	12.7	32.6	8.3	59.7	105	
	1.2	0.3	1.1	0.7	2.5	98	
Winter	50.2	19.9	49.9	15.5	96.6	42	< 0.001
	41.7	13.7	38.9	17.2	81.3	97	
	1.2	0.3	1.1	0.8	1.9	76	
Total	**32.9**	17.3	30.9	6.8	96.6	248	< 0.001
	26.3	14.8	24.0	5.4	81.3	512	
	1.4	0.6	1.2	0.5	4.5	418	

Notes: [a]The unit of ambient and personal PM$_{2.5}$ mass concentration is µg m^{-3}; [b]Ambient PM$_{2.5}$ concentrations obtained from seven EPD monitoring stations at different areas in Hong Kong; [c]SD refers to standard deviation; [d]N refers to the total number of samples; [e]The level of significance of mass difference was taken as $p < 0.05$.

Personal PM$_{2.5}$ concentrations in all the subjects ranged from 6.8 to 96.6 µg m^{-3} with an annual average of 32.9 µg m^{-3}. For personal PM$_{2.5}$ mass concentrations, the annual mean concentration was less than the HKEPD annual average air quality guideline of 35 µg m^{-3} (Figure 3-6). In contrast, personal PM$_{2.5}$ concentrations measured in other developed countries usually have considerably lower values than in Hong Kong, such as Baltimore (mean = 12.9 µg m^{-3}) (Williams et al., 2000) in the US, Toronto (mean = 22.0 µg m^{-3}) in Canada (Kim et al., 2005). The annual average and median ambient PM$_{2.5}$ concentrations were 26.3 ± 14.8 and 24.0 µg m^{-3}, respectively. The lowest ambient PM$_{2.5}$ was 5.4 µg m^{-3}, and the highest ambient concentration was 81.3 µg m^{-3} during the measurement period. The highest average ambient PM$_{2.5}$ was observed in winter (41.7 ±

13.7 µg m^{-3}), which was 2.9 times higher than those measured in summer (14.1 ± 5.6 µg m^{-3}). Average personal exposure to PM$_{2.5}$ demonstrated significant (p < 0.01) seasonal variation with the highest average concentration in winter (50.2 ± 19.9 µg m^{-3}) and the lowest level in summer (22.5 ± 11.8 µg m^{-3}). On a day-to-day basis, as shown in Table 3-11, the average personal PM$_{2.5}$ concentrations was typically found to be 6.6 µg m^{-3} (*p* = 0.009) greater than corresponding ambient PM$_{2.5}$. The P/A concentration ratios in different seasons were presented in Table 3-12. It was observed that the average P/A ratio ranged from 1.2 to 1.8 during different seasons.
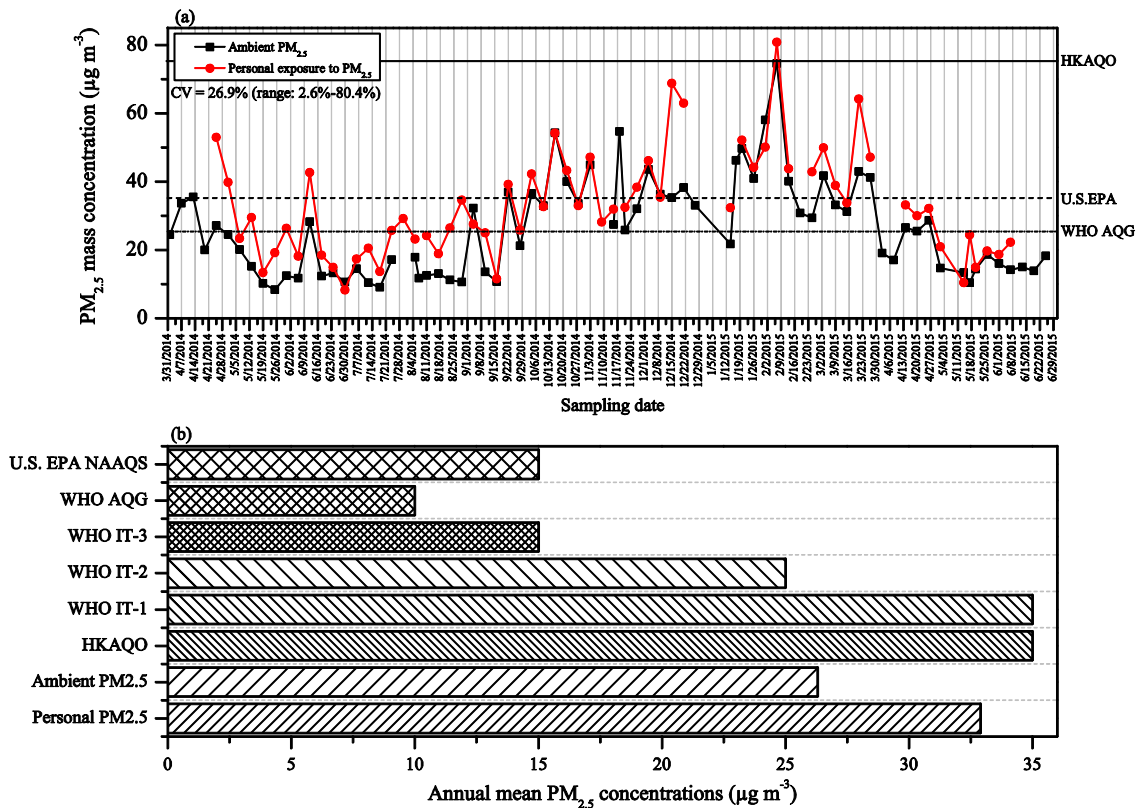

Figure 3-7. (a) Time Series Plots of Daily Average Ambient and Personal Exposure to PM$_{2.5}$ Throughout the Study Period and (b) annual mean PM$_{2.5}$ concentrations from ambient and personal samples in Hong Kong in comparison to those of WHO, U.S.EPA, and HKEPD.

As shown in Figure 3-6 (a), personal PM$_{2.5}$ demonstrated similar variation over time as ambient PM$_{2.5}$. Higher personal exposure levels were observed when compared to ambient concentrations for the one-year sampling campaign. In line with the findings of this study, similar results (personal exposures exceeded ambient/outdoor concentrations) were reported in other cities (Janssen et al., 1998; Jahn et al., 2013). A similar seasonal variation was found in Schedule A for all subjects with higher PM$_{2.5}$ exposures in winter

and lower in summer. Individual personal exposure concentrations higher than the ambient $PM_{2.5}$ were more than 80% of the samples days during the one-year sampling period. Daily average personal $PM_{2.5}$ concentrations exceeded the U.S. EPA 24-hr mean air quality guideline of 35 $\mu g\ m^{-3}$ in 36.5% of the sampling days. In addition, the WHO air quality guideline of 25 $\mu g\ m^{-3}$ was exceeded in 66.7% of the sampling days, most of these high exposure levels occurred in winter. Both ambient concentrations and personal exposure to $PM_{2.5}$ complied with the annual HKAQO and WHO IT-1 of 35 $\mu g\ m^{-3}$.
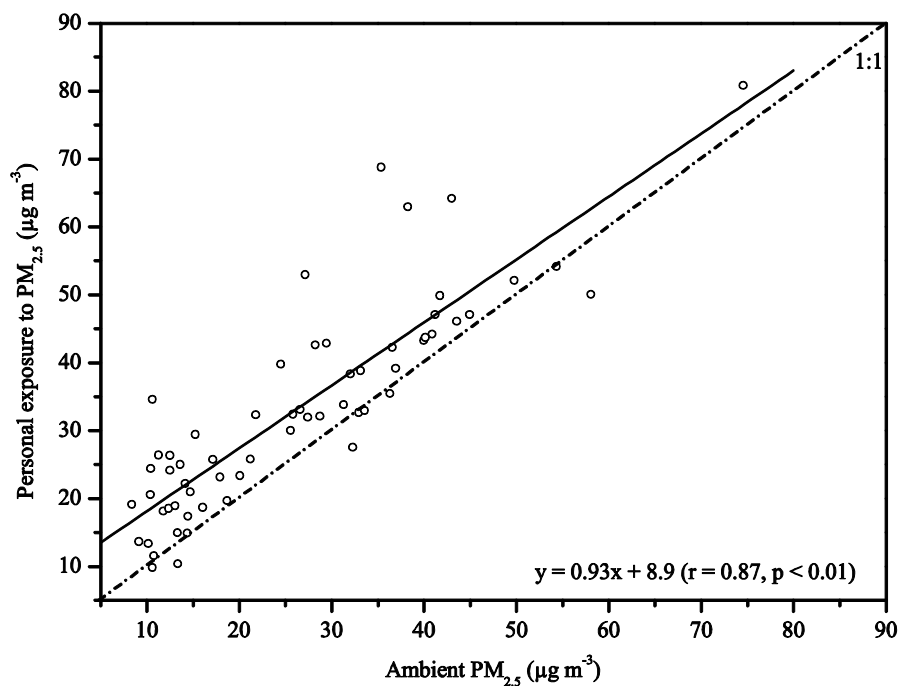

Figure 3-8. Correlation Between Daily Average Ambient and Personal Exposure to $PM_{2.5}$ Throughout the Sampling Period

Daily average personal $PM_{2.5}$ concentrations were highly correlated with ambient concentrations over time for all subjects, and significant positive correlation ($r = 0.87$, $p < 0.01$) was shown (Figure 3-7). A regression analysis ($P = \alpha \cdot A + N$) of the total personal exposure, P, against A yields a slope (α), and an intercept equal to the average non-ambient exposure (N) (Wilson et al., 2000). Non-ambient exposure, N, includes exposure to particles generated by indoor sources and particulate matter related to individuals' personal activity (Wilson and Brauer, 2006). The regression of individual P against A is

shown in Figure 3-8. The average personal exposure to non-ambient $PM_{2.5}$, estimated by the interception, is 8.80 µg m^{-3}, compared to 9.4 ± 6.9 µg m^{-3} from the average of the individual intercepts (Table 3-13). Based on the measured exposure, a mean $PM_{2.5}$ personal cloud of 3.1 µg m^{-3} was estimated in the Baltimore Particulate Matter Epidemiology-Exposure Study. An earlier analysis of $PM_{2.5}$ from a personal monitoring panel study in Vancouver, Canada found similar mean levels of non-ambient generated exposures (8.47 µg m^{-3}) (Wilson and Brauer, 2006). Non-ambient generated exposure (range: 5.0-6.4 µg m^{-3}) was estimated from a 6-week children's exposure study in Prince George, Canada (Noullett et al., 2010). The non-ambient component of total personal exposure refers to exposure to PM generated by indoor sources and individual's personal activities. The non-ambient generated exposure is independent of ambient concentration (Ebelt et al., 2005; Wilson and Brauer, 2006). As expected, the non-ambient exposure (or the mass difference between personal exposure and ambient concentration) was not related to the IAQ of Hong Kong.

Table 3-13. Personal Exposure to $PM_{2.5}$ for Each Subject, and Spearman's Correlation Between Ambient and Personal $PM_{2.5}$ by Subjects During the Study Period

Subject ID	Personal $PM_{2.5}$[b]	CV (%)	Personal vs. ambient			
			Spearman's Rho	Slope	Intercept[a]	Pairs (n)
G101	22.3 ± 11.2	50.2	0.87**	0.68	4.6	20
G102	29.2 ± 16.4	56.2	0.90**	1.02	4.6	20
G103	40.9 ± 18.7	45.7	0.59**	0.91	20.0	19
G104	26.0 ± 9.8	37.7	0.70**	0.62	8.9	19
G201	35.2 ± 23.8	67.6	0.82**	1.17	4.6	20
G202	33.3 ± 21.6	64.9	0.81**	1.13	3.7	21
G203	39.2 ± 21.5	54.8	0.74**	0.98	12.8	20
G204	30.9 ± 13.3	43.0	0.69**	0.70	12.9	19
G301	38.0 ± 12.6	33.2	0.44	0.59	22.8	19
G302	31.8 ± 13.4	42.1	0.80**	0.99	4.8	19
G303	29.5 ± 13.2	44.7	0.96**	1.08	1.0	21
G304	39.7 ± 18.3	46.1	0.73**	0.99	12.5	19
Mean $\pm$ SD[c]			0.75 ± 0.14	0.91 ± 0.20	9.4 ± 6.9	
Median			0.77	0.99	6.9	

Notes: [a]The unit of personal $PM_{2.5}$ is µg m^{-3}; [b]Values represent average $\pm$ standard deviation; [c]SD refers to standard deviation; **Correlation is significant at the 0.01 level (2-tailed); *Correlation is significant at the 0.05 level (2-tailed).

Summary statistics of individual $PM_{2.5}$ mass concentrations by subjects from all days of personal monitoring are shown in Table 3-12. Figure 3-6 and Table 3-12 also relate to the inter- variability and intra-variability in exposures observed between and

within the subjects. The variability of personal exposure between the days, expressed by the coefficient of variance between days ranged from 2.6% to 80.4% with an overall mean of 26.9% (Figure 3-6). Intra-individual variability for each subject during the study period ranged from 33.2% to 67.6% (Table 3-13).

As shown in Table 3-13, when comparing the personal $PM_{2.5}$ exposure levels, one student (subject G103) showed highest average concentration (40.9 ± 18.7 µg m^{-3}), and lowest (22.3 ± 11.2 µg m^{-3}) was found for the office worker (subject G101). In order to assess the relationship between ambient and personal exposure, correlation analysis was performed during each subjects' sampling period. As reported in Table 3-13, the association is influenced by the variation in personal exposures between subjects. Moderate to high correlations (range: 0.44-0.96) between ambient and personal $PM_{2.5}$ was shown. The median of the individual Spearman's r_s was 0.77 for all subjects. The differences in r_s may suggest that ambient concentration is an adequate surrogate for ambient-generated exposure but not for total personal exposure. The regression analysis was related to each subject's time activity profile to better understand the sources of exposure to $PM_{2.5}$. The individual intercepts ranged from 1.0 to 22.8 µg m^{-3}. According to the one-year activity diaries, the office worker subjects (e.g., G101, G104, G302) spent an average of 90.2% of each day in an indoor environment. The average daily time contribution for office workers spent on transportation and outdoor were equal to 4.6% and 5.1%, respectively. Lower intercepts and higher correlation coefficients were observed among office workers. For non-office workers (e.g., G203, G301, G304), they spent 71.3% of their daily time in indoors while a considerable portion of time was spent outdoors (21.3%) and on transportation (7.4%). For example, non-office worker subject G301, who spent 32.0% of daily time outdoors, 7.5% of the time on transportation. The lowest correlation coefficient and highest intercept were found for subject G301, suggesting that outdoor ambient $PM_{2.5}$ was not a strong predictor of personal exposure for this individual.
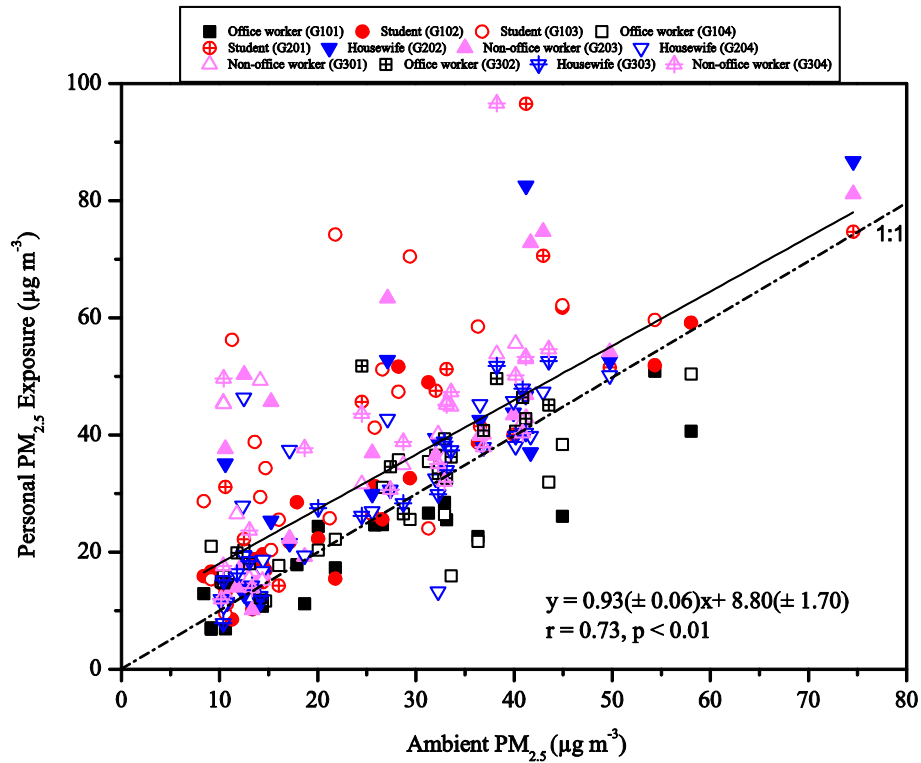
Figure 3-9. The Relationship Between the Individual Personal Exposure to $PM_{2.5}$ and the Ambient Concentrations of $PM_{2.5}$

As shown in Table 3-14, when comparing the personal $PM_{2.5}$ exposure levels from the occupation of different subjects during the one-year long measurement, the non-office worker subjects showed highest average concentrations (39.0 ± 17.9 µg m^{-3}) during the entire study period, while the lowest average concentrations were measured among the office workers (26.7 ± 12.0 µg m^{-3}). Consistent with the results illustrated in Schedule A, lowest exposure was observed compared to non-office workers in the one-year sampling period. The coefficients of variance were also shown in Table 3-13, and the personal $PM_{2.5}$ for non-office workers were slightly more variable than the office workers. As also shown in Table 3-14, the lowest P/A concentration ratios were observed among office worker with an average of 1.1 ± 0.4, and the highest P/A ratios were reported for non-office workers (1.6 ± 0.9).

Table 3-14. Statistical Description of Personal Exposure to PM$_{2.5}$ for Different Subjects

	Students	Office Workers	Housewives	Non-office workers
Mean[a]	35.0	**26.7**	31.2	**39.0**
SD[b]	20.1	12.0	16.4	17.9
Median	29.0	25.6	29.1	39.9
Minimum	8.5	6.8	7.8	10.1
Maximum	96.5	51.8	86.7	96.6
CV (%)[c]	57.5%	44.9%	52.5%	45.2%
N[d]	62	61	64	61
P/A ratio	1.5 ± 0.8	1.1 ± 0.4	1.3 ± 0.5	1.6 ± 0.9

Notes: [a]The unit of personal PM$_{2.5}$ mass concentration is µg m^{-3}; [b]SD refers to standard deviation; [c]refers to the coefficient of variance; [d]N refers to the number of samples.

Table 3-15 reported the concentrations of major elements in ambient and personal PM$_{2.5}$ samples. In addition, correlation coefficients (Spearman's r_s) and coefficient of divergence (COD) of elements in ambient and personal PM$_{2.5}$ were also listed in Table 3-15. To express the relationship between ambient and personal exposures, the personal-to-ambient (P/A) component ratio was calculated for each sampling day (Figure 10). The median concentrations of crustal elements (e.g., Ca, Ti, Fe) were higher in personal PM$_{2.5}$ (128.6, 10.9, 183.7 ng m^{-3}, respectively) than in ambient PM$_{2.5}$ (84.5, 8.8, 159.1 ng m^{-3}, respectively) (Table 3-15). However, moderate correlations were observed for Ca ($r_s = 0.59$), Ti ($r_s = 0.78$) and Fe ($r_s = 0.67$), respectively, in ambient and personal PM$_{2.5}$. The mean and median P/C ratios greater than unity were observed for Ca, Ti and Fe. The personal PM$_{2.5}$ samples were clearly enriched in crustal origin (Ca, Ti and Fe), indicating the contribution of indoor sources (e.g, cooking, secondhand smoke, resuspension) and/or personal activities (e.g., cleaning). This is in agreement with the findings of other studies for crustal particles (Hopke et al., 2003). Larson et al. (2004) reported that personal exposure to crustal sources were strongly associated with personal activities.

The concentrations of Al (156.7 ± 99.4 ng m^{-3}) and Si (263.3 ± 225.5 ng m^{-3}) were higher in ambient PM$_{2.5}$, significant correlations (r_s range: 0.77-0.85, $^{**}p < 0.01$) were observed in ambient and personal PM$_{2.5}$. Trace elements concentrations such as V, Mn, Ni, Cu, Zn, and Pb were slightly higher in ambient than in personal PM$_{2.5}$, and moderate to high correlation coefficients (r_s varied from 0.59 to 0.84) were observed for elements in ambient and personal PM$_{2.5}$ samples. Pb and Zn act as the marker elements for motor vehicle emissions, while Ni and V are primarily from the combustion of heavy fuel oil (Lai et al., 2007). As shown in Figure 10, Pb and Zn have P/A ratios >1, indicating the

dominance of ambient sources, the presence of indoor sources (e.g., secondhand smoke, cleaning products) and contribution of personal activities (e.g., transportation, along roadside).

In contrast, mean and median P/A ratios less than unity were observed for Ni and V, which indicated that these metals were more likely originated from ambient sources. Highest sulfur concentrations were observed in ambient (2878.0 ± 1698.5 ng m^{-3}) and personal PM$_{2.5}$ samples (2510.7 ± 1273.8 ng m^{-3}). Sulfur is a marker of aerosol which penetrates from outdoors, because of its limited indoor sources. The correlation between sulfur in ambient PM$_{2.5}$ and sulfur in personal PM$_{2.5}$ was statistically significant ($r_s = 0.95$, $p < 0.01$).

The coefficient of divergence (COD) analysis for elements (listed in Table 3-14) in personal and ambient PM$_{2.5}$ was calculated during the sampling campaign. If two samples (e.g., ambient, personal) are very similar, COD approaches zero, otherwise, COD approaches one. The lowest COD value was observed for sulfur (0.13), indicating that sulfur is uniformly distributed. Higher COD were observed for re-suspended dust (0.3-0.5), trace elements (0.5-0.6) (e.g., As, Cr), and NaCl factors (0.6).
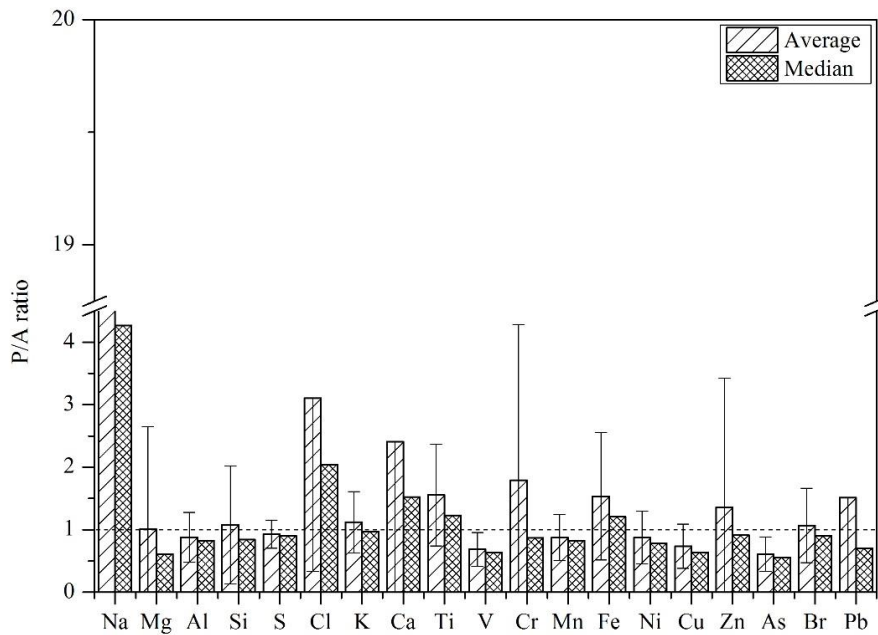
Figure 3-10. Mean (standard deviation) and median P/A ratio of elements.

      Our results are comparable to previous finding suggesting that ambient measurement may not be adequate to estimate personal levels. As suggested by Ozkaynak et al. (1996) in the PTEAM study, weak Pearson correlation (r = 0.4) between outdoor concentrations and personal PM10 exposure was observed, indicating only 16% of the variation in measured exposures was due to outdoor concentration. Wilson and Brauer (2006) reported that for a longitudinal panel studies an average of Pearson's r 0.36 was found between individuals' personal exposures and ambient concentrations. Jahn et al. (2013) found that less than half of the monitored subjects had moderate longitudinal correlations (Spearman's rs > 0.5) of personal exposure with ambient concentrations. Additional studies were carried out in the United States and Europe, with similar findings (Oglesby et al., 2000; Wallace, 2000; Williams et al., 2000).

Table 3-15. Summary of the Major Elements in Ambient and Personal $PM_{2.5}$

Component	Ambient (A, ng m^{-3})				Personal (P, ng m^{-3})				P-A	N (Pairs)	COD
	Mean ± SD[a]	Median	Minimum/Maximum	N[b]	Mean ± SD	Median	Minimum/Maximum	N	(Spearman's r_s)	Daily Average	
Na	1574.3 ± 2236.7	421.1	46.1/12409.1	510	3034.8 ± 1893.5	2489.3	103.2/9373.8	246	0.105	60	0.61
Mg	180.9 ± 96.4	170.3	3.0/816.6	373	127.8 ± 86.0	112.0	13.0/435.2	175	0.197	53	0.49
Al	156.7 ± 99.4	134.9	21.0/456.8	470	131.8 ± 99.0	123.8	7.1/619.5	234	0.770**	58	0.29
Si	263.3 ± 225.5	222.5	3.4/1047.4	482	255.8 ± 285.4	176.4	4.0/2412.2	235	0.849**	59	0.31
S	2878.0 ± 1698.5	2542.4	342.3/8578.7	512	2510.7 ± 1273.8	2321.3	84.8/6323.7	248	0.954**	60	0.13
Cl	128.6 ± 312.7	37.6	5.0/2893.1	478	176.2 ± 258.0	72.7	6.1/1632.7	245	0.740**	60	0.48
K	283.0 ± 224.3	200.7	23.2/1058.5	512	283.0 ± 225.0	235.1	20.6/1302.8	248	0.862**	60	0.21
Ca	95.9 ± 63.1	84.5	14.2/374.3	512	201.4 ± 654.3	128.6	17.5/10217.6	248	0.594**	60	0.34
Ti	10.5 ± 8.1	8.8	1.2/96.1	459	13.3 ± 10.0	10.9	1.4/64.2	239	0.777**	60	0.29
V	23.4 ± 28.4	14.2	1.3/218.2	507	15.6 ± 15.4	11.4	1.3/94.3	240	0.792**	60	0.32
Cr	3.1 ± 1.6	2.9	0.9/9.6	350	3.9 ± 3.9	2.6	0.9/35.0	187	0.229	47	0.55
Mn	13.0 ± 9.0	11.3	0.8/64.5	442	11.4 ± 8.9	10.2	0.9/56.0	227	0.844**	58	0.28
Fe	179.0 ± 107.0	159.1	12.2/574.2	512	231.4 ± 179.1	183.7	8.1/1161.8	248	0.670**	60	0.26
Ni	6.6 ± 7.1	4.2	0.6/56.6	497	5.6 ± 4.6	4.1	0.4/25.4	239	0.588**	59	0.26
Cu	19.7 ± 15.9	16.1	0.5/85.5	492	15.3 ± 20.3	10.6	0.7/250.1	243	0.796**	60	0.31
Zn	111.6 ± 111.3	66.6	1.3/805.8	495	105.5 ± 133.0	68.3	0.6/1366.2	248	0.821**	60	0.27
As	7.0 ± 6.3	5.4	0.8/35.4	195	3.0 ± 2.1	2.5	0.8/13.0	134	0.272	27	0.57
Br	12.6 ± 11.2	9.7	0.5/58.6	475	11.4 ± 10.9	9.3	0.7/87.6	246	0.870**	60	0.21
Pb	33.2 ± 32.1	22.0	1.4/161.3	402	25.1 ± 36.1	18.4	1.5/440.6	210	0.816**	51	0.11

Notes: [a]SD refers to standard deviation; [b]N refers to the number of valid data, and concentrations below the MDL have been discarded; **Correlation is significant at the 0.01 level (2-tailed).

4. SUMMARY AND RECOMMENDATIONS

A total of 167 PM$_{2.5}$ personal exposure samples were obtained during summer and winter of 2014 from more than 40 adult residents of Hong Kong in Schedule A. Seasonal variations of personal PM$_{2.5}$ exposures and their chemical species were investigated. Higher average personal exposures observed in winter than that in summer, and seasonal variations were significant ($p < 0.05$). The lowest personal exposures (e.g., PM$_{2.5}$ concentrations, carbonaceous aerosol, water-soluble ions) were reported among office workers during the study period. No significant gender or spatial difference was found for subjects residing in different areas in Hong Kong in the present study.

A total of 248 personal PM$_{2.5}$ samples were collected every eighteenth day by twelve adult subjects from April 2014 to June 2015 in Schedule B. Consisted with the results in Schedule A; personal exposures displayed significant seasonal variations with the lowest concentrations in summer and the highest in winter during the one-year long campaign. On a day-to-day basis, the average personal PM$_{2.5}$ was typically found to be 6.6 µg m^{-3} ($p < 0.01$) greater than the corresponding ambient PM$_{2.5}$ concentrations in this report. The correlation of personal exposures with ambient concentrations yielded values of Spearman r_s from 0.44 to 0.96 with an average of 0.77. The non-ambient generated exposure (intercept: 4.4-22.8 µg m^{-3}) is independent of ambient concentration, which refers to exposure generated by indoor sources (e.g., secondhand smoke, cooking, incense burning, cleaning) and individual's personal activities (e.g., transportation, along roadside) in this study.

Recommendations for future work:

Within-person and between-person variance:

The majority of the really high PM$_{2.5}$ personal exposures (e.g., subject G301, G103, G203) were not the result of high ambient concentrations, but were due to subjects' activities, and there was also a high degree of variability within and between subjects. Further investigation using more personal monitoring data from the time-microenvironment-activity diaries (e.g., location, activities) as independent variables in a

statistical model can provide important information regarding the sources of high exposures and possibly extend the information to subpopulations (e.g., office workers, high-exposure subjects).

Separate personal exposure into ambient and non-ambient exposure:

Personal $PM_{2.5}$ exposures were clearly impacted by both ambient and non-ambient sources. Ambient concentrations provide a useful surrogate for ambient-generated exposure but not for total exposure or non-ambient exposure. Stronger personal-ambient associations existed for sulfur compared to total $PM_{2.5}$ and other elements. The data analysis supports the use of sulfur and other chemical species (e.g., sulfate) as indicators of ambient generated exposure as well as develop methodologies (e.g., chemical tracers, ambient exposure factor) to differentiate personal exposure of ambient origin.

Identify and quantify sources of personal exposure to particles:

$PM_{2.5}$ exposures are impacted by ambient (outdoor), indoor sources and personal activities. The identification of different sources and their relative contribution to total personal exposures can provide valuable information for epidemiological studies and regulatory agencies.

5. REFERENCES

Adren, P. C., Burnett, R. T., Thurston, G. D., Thun, M. J., Calle, E. E. and co-authors. 2004. Cardiovascular Mortality and Long-Term Exposure to Particulate Air Pollution Epidemiological Evidence of General Pathophysiological Pathways of Disease. *Circulation* **109**, 71-77.

Analitis, A., Katsouyanni, K., Dimakopoulou, K., Samoli, E., Nikoloulopoulos, A. K. and co-authors. 2006. Short-term effects of ambient particles on cardiovascular and respiratory mortality. *Epidemiology* **17**, 230-233.

Beverland, I. J., Cohen, G. R., Heal, M. R., Carder, M., Yap, C. and co-authors. 2012. A comparison of short-term and long-term air pollution exposure associations with mortality in two cohorts in Scotland. *Environ Health Persp* **120**, 1281.

Cao, J., Lee, S., Ho, K., Zhang, X., Zou, S. and co-authors. 2003. Characteristics of carbonaceous aerosol in Pearl River Delta Region, China during 2001 winter period. *Atmos Environ* **37**, 1451-1460.

Cao, J. J., Lee, S. C., Chow, J. C., Cheng, Y., Ho, K. F. and co-authors. 2005. Indoor/outdoor relationships for PM2.5 and associated carbonaceous pollutants at residential homes in Hong Kong - case study. *Indoor Air* **15**, 197-204.

Chau, C., Tu, E. Y., Chan, D. and Burnett, J. 2002. Estimating the total exposure to air pollutants for different population age groups in Hong Kong. *Environ Int* **27**, 617-630.

Chow, J. C., Watson, J. G., Robles, J., Wang, X., Chen, L.-W. A. and co-authors. 2011. Quality assurance and quality control for thermal/optical analysis of aerosol samples for organic and elemental carbon. *Analytical and bioanalytical chemistry* **401**, 3141-3152.

Chow, J. C. and Watson, J. 2012. Chemical analyses of particle filter deposits. *Aerosols Handbook: Measurement, Dosimetry, and Health Effects* **2**, 177-202.

Chung, Y., Dominici, F., Wang, Y., Coull, B. A. and Bell, M. L. 2015. Associations between long-term exposure to chemical constituents of fine particulate matter (PM2. 5) and mortality in Medicare enrollees in the Eastern United States. *Environ Health Persp* **123**, 467.

Ebelt, S. T., Wilson, W. E. and Brauer, M. 2005. Exposure to ambient and nonambient components of particulate matter: a comparison of health effects. *Epidemiology* **16**, 396-405.

Ho, K., Cao, J., Harrison, R. M., Lee, S. and Bau, K. 2004. Indoor/outdoor relationships of organic carbon (OC) and elemental carbon (EC) in PM< sub> 2.5</sub> in roadside environment of Hong Kong. *Atmos Environ* **38**, 6327-6335.

Ho, K., Cao, J., Lee, S. and Chan, C. K. 2006a. Source apportionment of PM 2.5 in urban area of Hong Kong. *Journal of hazardous materials* **138**, 73-85.

Ho, K., Lee, S., Cao, J., Chow, J. C., Watson, J. G. and co-authors. 2006b. Seasonal variations and mass closure analysis of particulate matter in Hong Kong. *Sci Total Environ* **355**, 276-287.

Ho, K. F., Engling, G., Ho, S. S. H., Huang, R. J., Lai, S. C. and co-authors. 2014. Seasonal variations of anhydrosugars in PM2.5 in the Pearl River Delta Region, China. *Tellus Series B-Chemical and Physical Meteorology* **66**.

Hopke, P. K., Ramadan, Z., Paatero, P., Norris, G. A., Landis, M. S. and co-authors. 2003. Receptor modeling of ambient and personal exposure samples: 1998 Baltimore Particulate Matter Epidemiology-Exposure Study. *Atmos Environ* **37**, 3289-3302.

Jahn, H. J., Kraemer, A., Chen, X. C., Chan, C. Y., Engling, G. and co-authors. 2013. Ambient and personal PM2.5 exposure assessment in the Chinese megacity of Guangzhou. *Atmos Environ* **74**, 402-411.

Janssen, N. A. H., Hoek, G., Brunekreef, B., Harssema, H., Mensink, I. and co-authors. 1998. Personal sampling of particles in adults: Relation among personal, indoor, and outdoor air concentrations. *Am J Epidemiol* **147**, 537-547.

Ji, W. and Zhao, B. 2015. Estimating Mortality Derived from Indoor Exposure to Particles of Outdoor Origin.

Kim, D., Sass-Kortsak, A., Purdham, J. T., Dales, R. E. and Brook, J. R. 2005. Sources of personal exposure to fine particles in Toronto, Ontario, Canada. *J Air Waste Manag Assoc* **55**, 1134-1146.

Kim, K. H., Kabir, E. and Kabir, S. 2015. A review on the human health impact of airborne particulate matter. *Environ Int* **74**, 136-143.

Lai, S. C., Zou, S. C., Cao, J. J., Lee, S. C. and Ho, K. F. 2007. Characterizing ionic species in PM2.5 and PM10 in four Pearl River Delta cities, south China. *J Environ Sci (China)* **19**, 939-947.

Larson, T., Gould, T., Simpson, C., Liu, L. J. S., Claiborn, C. and co-authors. 2004. Source apportionment of indoor, outdoor, and personal PM2.5 in Seattle, Washington, using positive matrix factorization. *J Air Waste Manage* **54**, 1175-1187.

Louie, P. K., Chow, J. C., Chen, L.-W. A., Watson, J. G., Leung, G. and co-authors. 2005. PM 2.5 chemical composition in Hong Kong: Urban and regional variations. *Sci Total Environ* **338**, 267-281.

Noullett, M., Jackson, P. L. and Brauer, M. 2006. Winter measurements of children's personal exposure and ambient fine particle mass, sulphate and light absorbing components in a northern community. *Atmos Environ* **40**, 1971-1990.

Noullett, M., Jackson, P. L. and Brauer, M. 2010. Estimation and characterization of children's ambient generated exposure to PM2.5 using sulphate and elemental carbon as tracers. *Atmos Environ* **44**, 4629-4637.

Oglesby, L., Künzli, N., Röösli, M., Braun-Fahrländer, C., Mathys, P. and co-authors. 2000. Validity of ambient levels of fine particles as surrogate for personal exposure to outdoor air pollution—results of the European EXPOLIS-EAS Study (Swiss Center Basel). *J Air Waste Manage* **50**, 1251-1261.

Ozkaynak, H., Xue, J., Spengler, J., Wallace, L., Pellizzari, E. and co-authors. 1996. Personal exposure to airborne particles and metals: results from the Particle TEAM study in Riverside, California. *J Expo Anal Env Epid* **6**, 57-78.

Rohr, A. C. and Wyzga, R. E. 2012. Attributing health effects to individual particulate matter constituents. *Atmos Environ* **62**, 130-152.

Wallace, L. 2000. Correlations of personal exposure to particles with outdoor air measurements: A review of recent studies. *Aerosol Sci Tech* **32**, 15-25.

Watson, J., Lioy, P. and Mueller, P. 2001. The measurement process: Precision, accuracy, and validity. *Air Sampling Instruments for Evaluation of Atmospheric Contaminants* **8**.

WHO. 2012. Air quality and health. Fact sheet N°313. *World Health Organization*.

WHO. 2013. *Health Effects of Particulate Matter: Policy Implications for Countries in Eastern Europe, Caucasus and Central Asia*, World Health Organization Regional Office for Europe.

WHO. 2014. Burden of disease from ambient and household air pollution. In: *World Health Organization*.

Williams, R., Suggs, J., Creason, J., Rodes, C., Lawless, P. and co-authors. 2000. The 1998 Baltimore Particulate Matter Epidemiology-Exposure Study: part 2. Personal exposure assessment associated with an elderly study population. *J Expo Anal Environ Epidemiol* **10**, 533-543.

Wilson, W. E., Mage, D. T. and Grant, L. D. 2000. Estimating separately personal exposure to ambient and nonambient particulate matter for epidemiology and risk assessment: Why and how. *J Air Waste Manage* **50**, 1167-1183.

Wilson, W. E. and Brauer, M. 2006. Estimation of ambient and non-ambient components of particulate matter exposure from a personal monitoring panel study. *J Expo Sci Environ Epidemiol* **16**, 264-274.

APPENDIX