Title

Assessing and Reducing Los Angeles Taxi Driver Occupational Exposures to Particulate Matter and Polycyclic Aromatic Hydrocarbons

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Assessing and Reducing Los Angeles Taxi Driver Occupational Exposures to Particulate Matter and Polycyclic Aromatic Hydrocarbons

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ABSTRACT OF THE DISSERTATION

Assessing and Reducing Los Angeles Taxi Driver Occupational Exposures to Particulate Matter and Polycyclic Aromatic Hydrocarbons

by

Nu Yu
Doctor of Philosophy in Environmental Health Sciences
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Professor Yifang Zhu, Chair

Taxi drivers and passengers are exposed to traffic related air pollutants (TRAPs), but their exposures and mitigation strategies were rarely explored. In passenger vehicles, the particulate matter (PM) concentrations can be 10 times higher than those in the general ambient environment, and the taxi vehicles are usually leakier than regular passenger vehicles due to their more frequent use and greater wear and tear.

This study investigated fine ($PM_{2.5}$) and ultrafine particle (UFP) levels outside and inside taxis, and their in-cabin to on-road (I/O) ratios under four different ventilation and mitigation conditions. The taxi drivers’ pre- and post-test urine samples were collected and their urinary hydroxylated-polycyclic aromatic hydrocarbons (OH-PAHs) and malondialdehyde (MDA) were analyzed, as biomarkers for their PAH exposures and lipid peroxidation levels.

The four driving test conditions include realistic working (no mitigation; NM) condition, window closed (WC) condition, window closed and high efficiency cabin air (WC+HECA) filter condition, and HECA filter (HECA) condition. The results show that the average UFP and $PM_{2.5}$ levels inside taxis were 2.57 x 10^4 particles / cm^3 and 26 µg / m^3 under NM. The most stringent WC+HECA condition effectively reduced the in-cabin UFP and $PM_{2.5}$ and UFP by 47%, 37%, respectively, from the NM condition. Under the NM, the average in-cabin to on-roadway (I/O) ratios for UFP and $PM_{2.5}$ were 0.60 and 0.75 respectively. But when they were driven under WC+HECA, the average I/O ratios for UFP and $PM_{2.5}$ were significantly reduced to 0.47 and 0.52 respectively. Although reductions in PAH exposures were not significant, the post-test MDA concentrations were found to be significantly associated with the in-cabin $PM_{2.5}$ and UFP concentrations, suggesting the reduction of the drivers’ lipid peroxidation can be at least partially attributed to the $PM_{2.5}$ and UFP reduction by WC+HECA. Overall, these results suggest using HECA filters has the potential to reduce vehicle in-cabin particulate matter (PM) levels and protect drivers’ health.

The dissertation of Nu Yu is approved.

Shane Que Hee

Niklas Krause

Robert Weiss

Jun Wu

Yifang Zhu, Committee Chair

University of California Los Angeles

2017

To

my dear parents

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1. **Yu, N.,** Zhu, Y., Xie, X., Yan, C., Zhu, T., Zheng, M., *Characterization of Ultrafine Particles and Other Traffic Related Pollutants near Roadways in Beijing.* Aerosol and Air Quality Research, 2015. **15**(4): p. 1261-1269.
2. Shu, S., **Yu, N.,** Wang, Y., Zhu, Y., *Measuring and modeling air exchange rates inside taxi cabs in Los Angeles, California.* Atmospheric Environment, 2015. **122**: p. 628-635.
3. Lin, Y., Qiu, X., **Yu, N.,** Yang, Q., Araujo, J., Zhu, Y., *Urinary Metabolites of Polycyclic Aromatic Hydrocarbons and the Association with Lipid Peroxidation: A Biomarker-Based Study between Los Angeles and Beijing.* Environmental Science & Technology, 2016. **50**(7): p. 3738-3745

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PUBLICATIONS

1. **Yu, N.,** Shu, S., Zhang, L., Wu, J., Zhu, Y., *Modeling On-road Fine and Ultrafine Particle Concentrations in Los Angeles.* (in preparation)

2. **Yu, N.,** Shu, S., Lin, Y., Zhu, Y., *Assessing and Reducing Fine and Ultrafine Particles inside Los Angeles Taxis.* (in preparation)

3. **Yu, N.,** Shu, S., Lin, Y., She, J., Ip, H., Qiu, X., Zhu, Y., *Effects of High Efficiency Cabin Air Filter in Reducing Drivers' Roadway Particulate Matter and Polycyclic Aromatic Hydrocarbon Exposures and Lipid Peroxidation.* PLOS ONE (in revision)

4. **Yu, N.,** Zhu, Y., Xie, X., Yan, C., Zhu, T., Zheng, M., *Characterization of Ultrafine Particles and Other Traffic Related Pollutants near Roadways in Beijing.* Aerosol and Air Quality Research, 2015. **15**(4): p. 1261-1269.

5. Shu, S., **Yu, N.,** Wang, Y., Zhu, Y., *Measuring and modeling air exchange rates inside taxi cabs in Los Angeles, California.* Atmospheric Environment, 2015. **122**: p. 628-635.

6. Lin, Y., Qiu, X., **Yu, N.,** Yang, Q., Araujo, J., Zhu, Y., *Urinary Metabolites of Polycyclic Aromatic Hydrocarbons and the Association with Lipid Peroxidation: A Biomarker-Based Study between Los Angeles and Beijing.* Environmental Science & Technology, 2016. **50**(7): p. 3738-3745

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1. **Yu, N.,** Shu, S., Lin, Y., Zhu, Y., *Assessing and Reducing Fine and Ultrafine Particles inside Los Angeles Taxis.* AIHce annual conference, Seattle, WA, June 2017

2. **Yu, N.,** *Taxi Drivers' Urinary PAH Metabolites and Association with Lipid Peroxidation.* AIHce annual conference, Baltimore, MD, June 2016

3. **Yu, N.,** Shu, S., Zhu, Y., Lin, Y., *Los Angeles Taxi Drivers' Occupational Exposure to Polycyclic Aromatic Hydrocarbons: Biomarkers and Health.* SCERC annual conference, UCLA, September 2015

4. **Yu, N.,** Shu, S., Lin, Y., She, J., Ip, H., Qiu, X., Zhu, Y., *Biological Marker Analysis for Particulate Matter and Polycyclic Aromatic Hydrocarbon Exposure.* JRI conference, PKU, China, August 2014

5. **Yu, N.,** Zhu, Y., Xie, X., Yan, C., Zhu, T., Zheng, M., *Characterization of Ultrafine Particles and Other Traffic Related Pollutants near Roadways in Beijing.* AAAR annual conference, Portland, OR, Oct 2013.

1. INTRODUCTION

1.1 Organization of the dissertation

This dissertation contains five chapters. Chapter 1 summarizes the background and motivation of this dissertation work leading to the development of the three major objectives. In Chapter 2, detailed experimental design and sampling methods and on-road PM results are summarized. From the sampling observations and summarized data, factors affecting on-road PM concentrations are analyzed and formulated on-road PM models. In Chapter 3, the in-cabin PM data are summarized and factors affecting I/O ratios are analyzed. Exposures in taxi drivers are assessed under different mitigation conditions. The ventilation, window position and cabin filter effects, and the car model effects are analyzed in detail. In Chapter 4, the taxi driver urinary PAH metabolites and MDA levels are summarized. Taxi drivers' occupational exposures to PM and PAH are analyzed, and the associations with oxidative stress levels are investigated. Finally, Chapter 5 provides an overall summary and conclusions of this dissertation work.

1.2 Taxi driver exposures to traffic related polycyclic aromatic hydrocarbons and particulate matter

High concentrations of polycyclic aromatic hydrocarbons (PAHs) and particulate matter (PM), such as fine ($PM_{2.5}$, aerodynamic diameter ≤ 2.5 μm) and ultrafine particles (UFPs, diameter $\leq$ 100 nm), are usually observed on and near roadways [1-3]. UFPs originate primarily from traffic emissions in urban environments [4]. Roadway UFP concentration is usually an order of magnitude higher than it is in the urban background [5]. The on-road UFP concentration typically ranges from 10,000 to 50,000 particles / cm^3, whereas the ambient background is in the order of 1,000 to 5,000 particles / cm^3 [5]. A previous study found a total of 27 PAHs in the air sampled from a city street was 8.3 times higher than that in the air sampled from a rural area, indicating higher PAH levels in roadways than those in the general ambient environment [6].

Influenced by the factors such as spatial features, traffic / meteorological conditions and seasonal effects, the on-road UFP and $PM_{2.5}$ concentrations show high temporal and spatial variations [7]. The use of one or a few fixed site monitoring stations to represent the average concentrations in a large urban area is not appropriate when studying UFP and $PM_{2.5}$ levels in roadways [8]. A previous study showed that generalized additive models (GAMs) explain 71% and 55% of the variances of the on-road $PM_{2.5}$ and UFP concentrations in southern California, respectively [9]. Another study with data collected on 75-mile freeway and 33-mile arterial roads in Los Angeles shows that a multiple regression model was able to explain 60-70% of the on-road UFP concentration variances [2]. However, the limited sampling time and routes made the models insufficiently reflect both the seasonal and spatial variability of PM.

Vehicle in-cabin PM levels are usually found to be lower than on-road levels because of the different mechanisms of particle loss [3]. Both on-road concentrations and the in-cabin to on-road (I/O) ratios determine in-cabin exposures. Research studies conducted on regular passenger vehicles showed that the I/O ratio of particulate pollutants ranged from 0-0.4 when the vehicle was in in-cabin recirculation (RC), and from 0.6 to 1.0 when the vehicle was in outside air (OA) mode [10]. Previous studies have found that ventilation modes, driving speed, cabin filtration performance, surface deposition and vehicle penetration were significant determinants of UFP I/O ratios in passenger vehicles [3, 11]. Other factors include vehicle model and car age / mileage [11, 12]. However, the way these factors influence taxi vehicles, which have limited vehicle models and relatively higher age / mileage than regular passenger vehicles, is still poorly understood.

Since the RC mode causes exhaled carbon dioxide (CO_2) to accumulate rapidly in the vehicle cabin, the current automotive ventilation systems and cabin air filters cannot control both PM and CO_2 simultaneously while preventing the infiltration of on-road traffic related air pollutants (TRAPs). Under the OA mode, the automotive ventilation system supplies roadway air into the taxi cabin and filters airborne particles. Although modern vehicles are mostly equipped with a cabin air filter, the passenger vehicle in-cabin PM concentration can remain an order of magnitude higher than the urban background. Considering that taxis are usually more leaky than regular passenger vehicles due to their high mileages and the greater wear and tear, their in-cabin levels could be even higher than the passenger vehicles.

Exposures to TRAPs, such as freshly emitted PM and PAHs are of special concern for certain subpopulations, such as taxi drivers who spend more time in traffic than the general population. A report from the Los Angeles Department of Transportation showed that there were more than 4000 taxi drivers working in Los Angeles for 72 hours per week on average [13, 14]. They potentially have higher occupational exposure to TRAPs because their total exposure was mainly affected by time spent in vehicle and length-weighted traffic count [15]. Given their long work hours and the highly congested Los Angeles roadways, the daily exposures of taxi drivers to traffic emitted PM are likely to be much higher than people in other occupations [16, 17]. This study evaluated the taxi drivers' PM and PAH exposures and the high efficiency cabin air (HECA) filter effects in taxis in Los Angeles.

1.3 Background and motivation

Air pollution is one of the leading risk factors of human premature death globally [18, 19]. As the most substantial combustion source in urban areas with dense population, vehicular emissions significantly increase carbon monoxide (CO), nitrogen oxides (NOx), $PM_{2.5}$, UFPs, and PAH concentrations on or near roadways and inside vehicles [3, 20-22]. Over 600 million people worldwide are exposed to these hazardous TRAPs [23, 24].

Among the TRAPs, UFP, $PM_{2.5,}$ and PAHs are widely found to be associated with various adverse health effects, such as respiratory and cardiovascular diseases, and premature mortality [25-27]. The International Agency for Research on Cancer (IARC) Working Group unanimously classified outdoor air pollution and PM as carcinogenic to humans (IARC Group 1) [28]. Freshly emitted UFPs from vehicle tailpipes are particularly rich in redox-active chemicals that lead to systemic inflammation [29, 30]. The on-road PM has been shown to induce oxidative stress, mitochondria damage, and acute pulmonary inflammation [31-33].

There is strong evidence to show that $PM_{2.5}$ and UFPs are more toxic per unit mass than larger particles because they can penetrate deeply into the lung and cross the 0.1-20μm thick lining cells [34], [35]. Some *in vitro* and *in vivo* studies also suggest that PAHs and their metabolites join the redox-active cycle and produce reactive oxygen species (ROS), which can in turn attack larger biological molecules including polyunsaturated lipids and form malondialdehyde (MDA) [36, 37]. The products of the peroxidation reactions have been demonstrated to be involved in the onset and development of a series of adverse health effects including cardiovascular diseases, diabetes, and cancers [38]. Since MDA is generated from lipid peroxidation, it is a widely used biomarker for evaluation of oxidative stress and related health effects [39, 40]. However, the association between PM / PAH exposures and lipid peroxidation showed inconsistency in previous studies, suggesting complex metabolizing processes [41, 42].

Due to high in-cabin levels of PM and PAHs, previous studies have shown that commuting exposure alone may count for up to 45-50% of the total daily exposure to UFPs in spite of the short average commuting time for regular passenger vehicle drivers (i.e. 1.3 hr / day). An occupational health study found that the lung cancer odds ratio among Canadian taxi drivers was 2.88 [43]. Another study on Turkish taxi drivers found cytogenetic damage in their peripheral lymphocytes [44]. A study of Chinese taxi drivers found heart rate variability (HRV) changes when the airborne $PM_{2.5}$ metallic components increased [45]. Considering the Los Angeles taxi drivers' long work hours and their leaky vehicles, the investigation of their exposure levels and the mitigation strategies is of urgent need.

1.4 Knowledge gap and research objectives

Because PM emitted from motor vehicles are primary pollutants that are rapidly transformed through physicochemical processes, which includes dispersion, coagulation, deposition, etc., the on-road PM concentration profiles show a very high temporal and spatial variation [7]. On-road PM models have been developed by air pollution researchers. However, previous studies indicate significant seasonal and spatial PM concentration and size distribution variations in southern California [46, 47], but the limited sampling time and routes of previously developed models insufficiently reflect both seasonal and spatial variability. The information gap in seasonal on-road PM variations and the limited data on surface streets made it difficult to fully explain the temporal and spatial variations of on-road PM concentrations in Los Angeles, and the PM concentration profile for different types of roads is important for evaluating human exposure to traffic related PM and the associated health risks.

Vehicle model and car age / mileage affect the PM I/O ratios [11, 12]. Other significant determinants of I/O ratios in passenger vehicles were ventilation modes, driving speed, cabin filtration performance, surface deposition and vehicle penetration [3, 11]. Setting the vehicle ventilation to outside air (OA) mode and improving cabin filter efficiency is a promising mitigation strategy to reduce in-cabin UFP and $PM_{2.5}$ and avoid in-cabin CO_2 accumulation. Our previous studies have shown that high efficiency cabin air (HECA) filters reduced in-cabin UFPs and $PM_{2.5}$ concentrations in passenger vehicles and school buses significantly under OA mode when windows were kept closed [48, 49]. However, taxi vehicles were found to be more frequently used than passenger vehicles with higher air exchange rates (AERs) [12]. Thus, the effectiveness of these mitigation strategies in reducing PM inside taxis needs to be further evaluated.

The fundamental goal of this research was to assess and reduce taxi driver exposures to on-road PM and PAHs. The objectives included: (1) Examining associations between on-road PM and predictive variables (traffic, road type, meteorology and spatial features) and modeling on-road PM concentrations in Los Angeles freeways, arterial roads and local surface streets. The models developed in this study can be combined with taxi vehicle I/O ratios for driver and passenger exposure quantification; (2) Identifying factors affecting the I/O ratios, and evaluate the effectiveness of various exposure mitigation strategies for taxis; (3) Assessing exposures to PM and PAHs among taxi drivers in Los Angeles and the effectiveness of closing windows and using HECA filters on PM and PAH exposure mitigation, and investigating the association among PM exposure, urinary PAH metabolites, and lipid peroxidation.

1.5 Future work

The roadway PM models and the temporal and spatial variance structures need to be further investigated because they don't explain more than 30% of the variance on freeways and local streets. The taxi driving speed effects on the I/O ratios are not linear and further investigation of the associations need to be done in the future. Taxi characteristic information, such as mile per gallon of fuel and total mileage of vehicle effects on the external PM and PAH concentrations will be analyzed.

2 ON-ROAD UFP AND $PM_{2.5}$ MEASUREMENTS AND MODELING

2.1 Abstract

Air pollution is among the top threats to public health in Los Angeles, and traffic related particulate matter (PM) exposure has been linked to different adverse health effects, such as respiratory and cardiovascular diseases. In this study, fine ($PM_{2.5}$) and ultrafine particle (UFP) concentrations were measured on roadways outside moving taxi vehicles in the Greater Los Angeles area for 76 days from April to November 2013. Multiple linear regression (MLR) models, MLR models with temporal variance structure (sMLR models), generalized additive models (GAMs) and GAMs with temporal variance structures (sGAMs) were developed separately to fit the data collected from freeways, arterial roads, and local surface streets. Predictive variables were selected from the meteorological condition, traffic condition and spatial feature panels with backward elimination procedure. The results show that GAMs explain higher percentages of the total variance of both UFP and $PM_{2.5}$ concentrations than other models. The models perform better on the arterial road data than the freeway and local surface street data. The MLR models generated the best cross validation (CV) results with the least discrepancy between the general and CV R^2s. The modeling results show different relationships on different roadway types between the UFP and $PM_{2.5}$ outcomes and the selected predictors. Some of the predictors have inconsistent effects on on-road UFP and $PM_{2.5}$ in different roadways.

2.2 Introduction

Fine particles ($PM_{2.5}$) refer to the particles with 2.5 µm or smaller aerodynamic diameters, and are usually measured in mass concentration (mass per volume of air, i.e., µg / m^3). Ultrafine particles (UFP) refer to particles with 0.1 µm or smaller aerodynamic diameters, and are usually measured in number concentration (the number of particle per volume of air, i.e., counts / cm^3). $PM_{2.5}$ and UFPs are emitted during combustion processes, and vehicular emissions usually constitute the dominant source in an urban environment [50]. The $PM_{2.5}$ and UFP concentrations on and near urban roadways can reach very high levels such as 10 times higher than ambient concentrations. Traffic related air pollutants (TRAPs) such as the freshly emitted particulate matter (PM) contribute significantly to total personal UFP and $PM_{2.5}$ exposures in an urban environment such as Los Angeles [2, 15].

By definition, UFPs are included in $PM_{2.5}$ but are not the major part, because they are relatively small in size and mass. $PM_{2.5}$ is usually considered as a regional pollutant whereas the UFP is considered as a local pollutant, mainly because the $PM_{2.5}$ life time is longer than the UFP life time. The on-road UFPs are mainly emitted from motor vehicles as a primary pollutant, and rapidly transformed through the physicochemical processes, which includes dispersion, coagulation, deposition, etc. On-road $PM_{2.5}$ comes from the secondary formation of the freshly emitted air pollutants, vehicle tire / break wear, and the regional sources such as industrial and residential emission sources.

Vehicle exhaust undergoes two dilution stages after emission. During the *tailpipe-to-road* stage, dilution is dominated by traffic generated turbulence. During the *road-to-ambient* stage, dilution is mainly dependent on atmospheric turbulence. Thus on-road UFP and $PM_{2.5}$ concentration profiles show very high temporal and spatial variations [7]. Influencing factors include spatial features, traffic flow and meteorological conditions. The use of one or a few fixed site monitoring stations to represent the average concentration in a large urban area is usual for other pollutants, but is not sufficient when studying UFP and $PM_{2.5}$ levels in roadways [8].

Some stochastic on-road $PM_{2.5}$ and UFP models have been developed by air pollution researchers. A previous study showed that generalized additive models (GAMs) could explain 71% and 55% of the variance of on-road $PM_{2.5}$ and UFP concentrations in southern California, respectively [9]. The study data were collected during a 20 day (March to June, 2011) sampling campaign, and primarily on freeways (75% of the 210 mile sampling route). Another study with data collected on freeways and arterial roads in Los Angeles showed that a MLR model was able to explain 60-70% of the on-road UFP concentration variance [2]. However, other studies have indicated significant seasonal and spatial UFP concentration and size distribution variations in southern California [46, 47], but the limited sampling time and routes have made the previously developed models insufficiently reflect both the seasonal and spatial variability.

In addition, the Final Report for Arterial Speed Study to Southern California Association of Governments indicates that the total directional mileage ratio of freeways to arterial streets in southern California is 1:9 [51]. Due to heavy traffic congestion in Los Angeles area, direction services such as Google Map or WAZE are more likely to route drivers into arterial roads and local surface streets than to major freeways. Driving speed variability is higher on arterial roads than that on freeways, and the UFP and $PM_{2.5}$ concentrations on arterial roads and local surface streets are mainly affected by proximity to gasoline-powered vehicles undergoing hard accelerations [2]. This speed variability makes the vehicular emission rates on arterial roads and surface streets less consistent compared to the emission rates on freeways [52]. In addition, the most vulnerable resident groups such as children and elderly people tend to rely more on local street commuting than the healthy adults do. Therefore, different UFP and $PM_{2.5}$ concentration profiles for freeways, arterial roads and local streets would be very important for evaluating human exposure to freshly emitted UFP and $PM_{2.5}$ and the associated health risks.

The information gap in seasonal and spatial UFP and $PM_{2.5}$ variation, and the limited data for arterial roads and local streets mean that existing models cannot fully account for temporal and spatial variations of on-road $PM_{2.5}$ and UFP concentrations in all roadways. The hypothesis of study is that the on-road UFP and $PM_{2.5}$ variations can be explained by time of day, meteorological/traffic conditions, and spatial features. Thus, this research aimed to (i) model the on-road PM concentrations on Los Angeles freeways, arterial roads and local surface streets, (ii) compare the performance of different models, such as multiple linear regression (MLR) models, generalized additive models (GAMs), and GAMs with spatial variance structure (sGAMs), on predicting on-road $PM_{2.5}$ and UFP concentrations, and (iii) examine the association between on-road UFP and $PM_{2.5}$ and predictive variables.

2.3 Methods

2.3.1 On-road sampling

On-road $PM_{2.5}$ and UFP concentration measurements were collected from outside moving taxis from April 29 to November 7, 2013. The study region in the Greater Los Angeles area has complex roadway networks and high levels of traffic congestion. The sampling routes include major commuter and truck transport freeways, arterial roads, and local surface streets (Figure 2.1). The total distance driven by the 22 taxis in 76 calendar days was approximately 11,000 kilometers and the total hours of field measurement was approximately 500 hours.

A TSI 3007 Portable Condensation Particle Counter (CPC3007, TSI Inc., St. Paul, MN) was used to measure the UFPs with sizes range of 0.01 to 1.0 µm and concentration range of 0 to 10^5 particles / cm^3. A length of TSI conductive tubing (to prevent particle loss in the tubing due to electrostatic charge) was used to connect the sampling inlet on the instrument through the vehicle back windows to the outside roadway environment. A DustTrak (Model 8520, TSI Inc., St. Paul, MN) with a $PM_{2.5}$ impactor was deployed to measure the $PM_{2.5}$ concentrations with the same length of tubing extending outside the back windows. Both CPC and DustTrak are real-time direct reading instruments with data logging functions. The UFP and $PM_{2.5}$ concentrations were logged at 1-sec intervals during the experiments. On-road air temperature and relative humidity were collected simultaneously with the UFP and $PM_{2.5}$ measurements at 1-sec intervals with a TSI Q-Trak Plus Indoor Air Quality Monitor (Q-Trak Plus 8554, TSI Inc., St. Paul, MN). All sampling instrument was synchronized to local time manually before sampling. A portable BT-Q1000x Global Positioning System (GPS) data logger (QStartz, Taiwan) was placed in the sampling vehicle to record travelling routes and driving speeds. Specifically, the instant longitude, latitude, altitude and vehicle speed was recorded in every 15 seconds by the GPS. The GPS was synchronized automatically with local time.

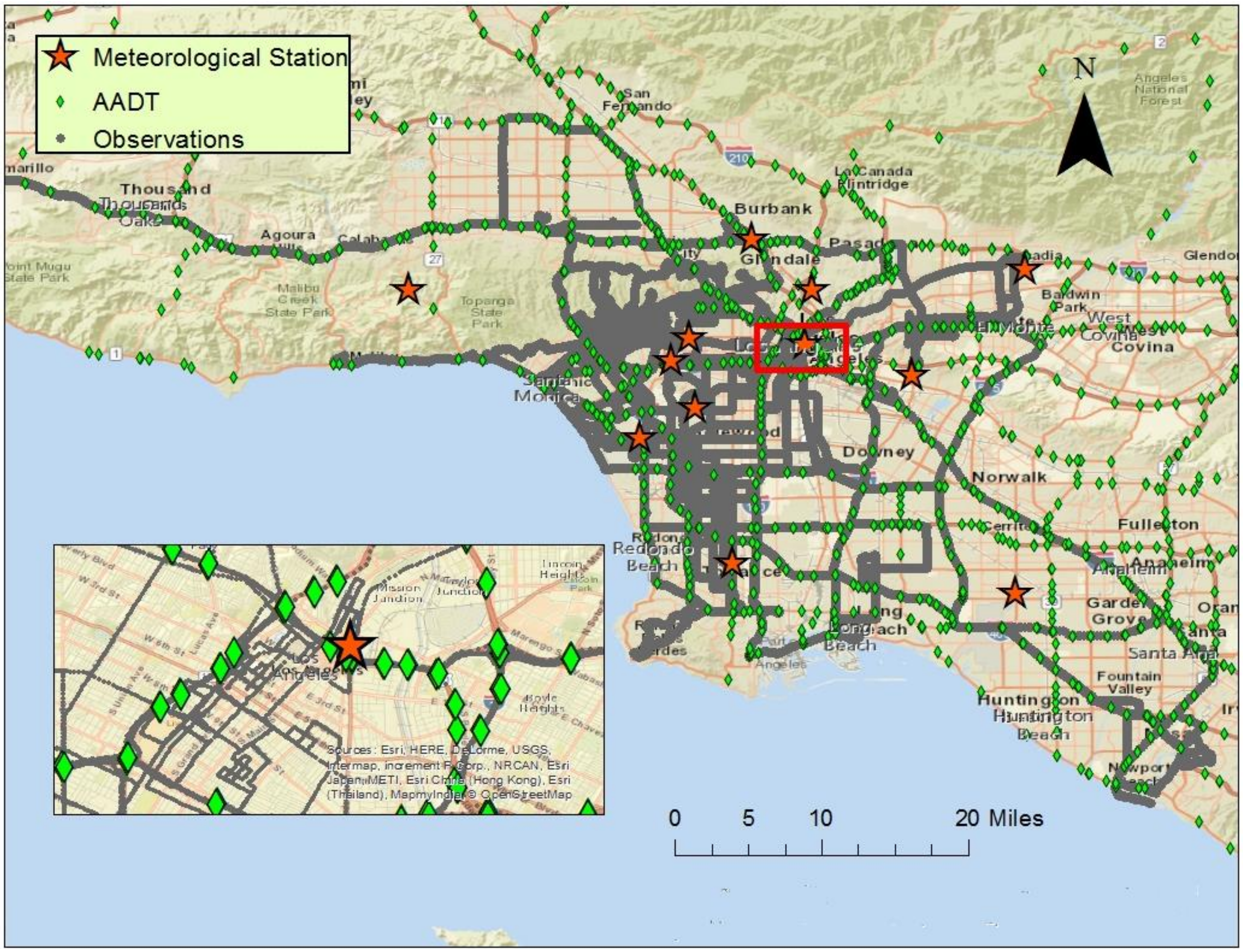


Figure 2.1. Map of the sampling routes, meteorological stations, and annual average daily traffic (AADT) data points in the Greater Los Angeles area. The sampled roadways are highlighted in gray. AADT data points are showed in green dots. The meteorological stations are showed in red stars.

2.3.2 Predictive variable compilation

A total of 13 predictive variables were grouped into three panels based on their physical properties. The meteorological parameter panel includes the on-road temperature / relative humidity, ambient temperature / relative humidity, and wind speed / direction (north-south wind speed and east-west wind speed). On-road temperature and relative humidity measurements were collected during the sampling campaign, at the same time with the target pollutant measurements. Ambient meteorological information were downloaded from a total of 12 monitoring stations of the Weather Underground sites (www.wunderground.com) in the Greater Los Angeles area (Figure 2.1), where the ambient air temperature, relative humidity, wind speed and wind directions were recorded in 15-minute intervals. When the data were compiled, each air pollutant observation was matched to the ambient temperature, relative humidity, wind speed and direction from the nearest station, with the nearest time stamp. The on-road temperature and humidity sampling readings show correlations with the nearest Weather Underground site data (Pearson's correlation R = 0.80). Thus each final model includes only on-road or ambient meteorological variables. Sampling season is also included in the meteorological predictor panel. This variable includes warmer and colder seasons. July to October was defined as the warmer season, and the colder season includes all other months.

The panel of traffic predictors includes the annual average daily traffic (AADT), vehicle speed, traffic static or moving, and weekday/weekend. The 2013 AADT counts were retrieved from the California Department of Transportation (CalTrans) Performance Measurement System (PeMS). However, the AADT data compiled from the PeMS database were only available for freeways and arterial roads in Los Angeles. Traffic static or moving (static) was set as a dichotomous variable based on whether the vehicle speed was less than 1 km / hr (coded as 1 when speed < 1 km / hr). Since the information of “static” variable was included in the “speed” variable, only one of them was included in the final models as predictors. Weekday is also a dichotomous variable that describes weather the on-road samples were collected during a regular weekday (coded as 1) or weekend (coded as 0).

The spatial feature panel includes the land use information and altitude. The land use information was obtained from the Topological Integrated Geographic Encoding and Referencing (TIGER) Products maintained by the United States Census Bureau. The retrieved land use features were classified into industrial, residential, retail and others categories. Altitude information was recorded by the GPS inside the sampling vehicle.

All predictors are summarized in Table 2.1.

Table 2.1. List of all predictive variables

Predictor	**Data source**
time of day	GPS (QSTARTZ BT-Q1000x)
Panel 1: meteorological conditions	
On-road temperature	On-road sampling with TSI Q-Trak (TSI 8554)
On-road relative humidity	On-road sampling with TSI Q-Trak (TSI 8554)
Ambient temperature	Nearest meteorology station data (www.wunderground.com)
Ambient relative humidity	Nearest meteorology station data (www.wunderground.com)
North-south wind speed	Calculated from the nearest meteorology station data (www.wunderground.com); north-positive, south-negative
East-west wind speed	Calculated from the nearest meteorology station data (www.wunderground.com); east-positive, west-negative
Season	Warmer: July, August, September, and October (coded as 1) Colder: April, May, June, and November (coded as 0)
Panel 2: traffic conditions	
Annual average daily traffic (AADT)	California Department of Transportation (CalTrans) Performance Measurement System (PeMS)
Vehicle speed	On-road sampling with GPS (QSTARTZ BT-Q1000x)
Static	On-road sampling (coded as 1 when speed < 1 km / hr; coded as 0 when speed ≥ 1 km / hr)
Weekday/weekend	Weekday: Monday thru Friday (coded as 1) Weekend: Saturday and Sunday (coded as 0)
Panel 3: Spatial features	
Land use	Topological Integrated Geographic Encoding and Referencing (TIGER) Products maintained by the United States Census Bureau
Altitude	On-road sampling with GPS (QSTARTZ BT-Q1000x)

2.3.3 Modeling

Before model development, dummy variables were set up for categorical predictors such as land use, static, weekday, static, and season. 1-min averages were calculated from the 1-sec on-road UFP and $PM_{2.5}$ measurements. A base 2 log transformation was used to reduce the skewness of the UFP and $PM_{2.5}$ sampling result distributions.

The MLR models describe the linear relationships between the predictors and outcome variables. They have been widely used in predicting air pollutant levels [53]. During the MLR model development, all available predictive variables were put into the models at the initial step, then a backward elimination method was used to select variables based on the calculated total model R^2s and Bayesian information criteria (BICs). Under this approach, the least significant variable was dropped at each step, while the calculated BIC change equals to or over 5, and there is no significant deterioration of the model fit (general R^2 doesn't drop over 0.01). The process was repeated until no more variables can be deleted.

The model developed in this study can be illustrated in Equation 2.1.

$$y_t = \alpha_0 + \sum_{j=1}^{p} \alpha_j \, x_{jt} + \varepsilon_t \tag{2.1}$$

Where α_0 is the intercept, and α_js are the coefficients of each predictor, j is the index of selected predictors, y_t is the outcome variable at time t, and x_{jt}s are the predictors. ε_t is the error term.

The multivariable GAMs incorporate both linear and nonlinear relationships. Assuming the estimate of the expected on-road concentrations at time t is y_t , the functions $f_i\ (x),\ f_j\ (x)$ are fitted to satisfy equation 2.2.

$$y_t = \beta_0 + \sum_{i=1}^{q} f_i\left(x_t^i\right) + \sum_{j=q+1}^{q+p} f_j\left(x_{a_t}^j, x_{b_t}^j\right) + \sum_{k=q+p+1}^{q+p+m} \beta_k\, x_t^k + \sum_{l=q+p+m+1}^{q+p+m+n} factor(x_t^l) + \varepsilon_t \qquad (2.2)$$

where β_0 is the model's intercept; x_{a_t}(e.g. temperature or north-south wind speed) and x_{b_t}(e.g. relative humidity or east-west wind speed) represent on-road or ambient meteorological conditions at t; and x_t^i, x_t^k, and x_t^l are other independent variables, among which x_t^i are q continuous nonlinear variables, x_t^k are m continuous linear variables, and x_t^l are n categorical variables as factors. f_i and f_j are the smooth functions used to construct nonlinear relationships. Factor functions were used to transform dummy variables with their differential intercept coefficients solved. Similar as the MLR modeling, predictive variables were selected by the backward elimination procedure, total R^{2}s and BICs were calculated and compared to find the best fit.

The temporal GAMs (sGAMs) were developed with autoregressive structures to measure temporal autocorrelations based on Equation 2.3, in which ε_t includes serially correlated errors and is not negligible. The autocorrelated regression errors can be described with a term for the first-order autoregressive process, AR(1):

$$\varepsilon_t = \emptyset\varepsilon_{t-1} + \nu_t \qquad (2.3)$$

Where the “random shocks” ν_t are assumed to be Gaussian white noise, and $\nu_t \sim N\ (0, \sigma)$. Same as the GAMs, predictive variables were selected by the backward elimination procedure, model total R^2s of the predictors and BICs were calculated and compared to find the best fit.

2.3.4 Model validation

A 10 times × 10 fold cross validation (CV) procedure was used to validate the developed models [9]. The sampling data on each day were evenly divided into 10 segments by time. One segment was randomly selected as the test data, and the training data came from the nine unselected segments with the constraint that there was an interval of at least 7 minutes between the measurement time of the training samples and test samples to avoid autocorrelations. This was repeated ten times to generate the CV results.

2.4 Results and discussion

2.4.1 Outcome variables: UFP and $PM_{2.5}$

The roadways were classified into three types (i.e., freeway, arterial road, and surface street) based on the information retrieved from TIGER Products. One minute averages of the logged UFP and $PM_{2.5}$ readings were calculated. Sample sizes, means and interquartile ranges of UFP and $PM_{2.5}$ measurements on different roadways are presented in Table 2.2. The highest average on-road UFP and $PM_{2.5}$ concentrations were observed on freeways, which were 4.42×10^4 particles / cm^3 and 36 μg / m^3 respectively. The lowest average UFP concentration was found on local streets, which was just about 50% of the average concentration on freeways. However, the

$PM_{2.5}$ means were very similar on freeways and local streets (36 vs. 35 μg/m^3), and the lowest $PM_{2.5}$ mean concentration was found on arterial roads (31 μg/m^3).

Table 2.2. Summary of outcome variables (calculated from 1-minute averages)

Roadway Type	Freeway	Arterial Road	Surface Street
Sample size, (total data points) (% of missing data)	27,038 (6.8%)	21,749 (3.4%)	142,039 (8.5%)
UFP, mean (interquartile range) (x10^4 *particles/cm*^{3})	4.42 (2.38, 5.42)	2.31 (0.53, 3.03)	2.22 (1.13, 2.59)
$PM_{2.5}$, mean (interquartile range) (*μg/m*^{3})	36 (22, 43)	31 (17, 38)	35 (18, 43)

On-road vehicle emissions consist of a complex mixture of particulate and gaseous pollutants such as UFP, $PM_{2.5}$, volatile organic compounds (VOCs), nitrogen oxides (NOx), and carbon monoxide (CO). UFPs come from the engine's incomplete combustion, and sometimes secondary formation processes occur such as nucleation from other gaseous pollutants and photochemical reactions. The on-road sources of $PM_{2.5}$ are mainly the vehicle tire / brake wear ("abrasion") and the secondary formation such as the coagulation of the nuclei mode UFPs. Also, due to their size and chemical composition differences, $PM_{2.5}$ is more physically and chemically stable in the atmospheric environment than the UFPs, thus the $PM_{2.5}$ can travel longer distance from industrial or residential emission sources in addition to the local on-road traffic emission sources. Therefore, the on-road $PM_{2.5}$ concentrations were affected more by the background levels and showed higher consistency among different road types than the on-road UFP concentrations. Previous studies also show that the UFP concentrations correlate poorly with $PM_{2.5}$ in the same ambient air samples [54, 55], indicating the possibility of different sources.

Based on the sampling results in this study, models were developed for on-road $PM_{2.5}$ and UFP separately for different road types.

2.4.2 Model development

The MLR models, MLR models with AR(1) variance structure (sMLR models), GAMs, and GAMs with AR(1) variance structure (sGAMs) were developed to fit the measured on-road UFP and $PM_{2.5}$ concentrations. The four types of models perform differently among the three road type measurements. The R^2s of the four types of models were calculated as the total R^2s of all linear and non-linear terms included in the final model. The resulting model total general and cross validation (CV) R^2s are summarized in Table 2.3. The modeling results are bolded in Table 2.3 when both of the general and CV R^2s are above 0.30, indicating the models are reasonable and useful for estimating the roadway UFP and $PM_{2.5}$.

As Table 2.3 shows, for both UFP and $PM_{2.5}$, all four types of models performed well on the measurements collected from arterial roads. The general R^2 are all equivalent or above 0.40 with CV R^2s ranging from 0.34 to 0.73. But among the freeway and local street models, only the local street $PM_{2.5}$ sMLR model and GAM generated general and CV R^2s higher than 0.30 at the same time. All models fit freeway and local streets UFP measurements very poorly, with general R^2s ranging from 0.15 to 0.26 and CV R^2s ranging from 0.07 to 0.17. With the same dataset, the GAMs generated higher R^2s than the MLR models did, but the MLR models generated the least discrepancies between the general and CV R^2s (Table 2.3). These results indicate that the predictors used in these models are probably not the best ones for estimating freeway and local street UFP concentrations, or new models need to be developed.

Table 2.3. Model performance summary (general R^2 / CV R^2)

Outcome	**Roadway Type**	**MLR**	**sMLR**	**GAM**	**sGAM**
UFP	Freeway	0.20 / 0.17	0.15 / 0.07	0.26 / 0.11	0.22 / 0.15
	Arterial Roads	**0.55 / 0.49**	**0.63 / 0.50**	**0.74 / 0.54**	**0.49 / 0.39**
	Local Streets	0.14 / 0.10	0.19 / 0.11	0.28 / 0.16	0.20 / 0.18
$PM_{2.5}$	Freeway	0.34 / 0.22	0.44 / 0.21	0.50 / 0.22	0.24 / 0.19
	Arterial Roads	**0.82 / 0.73**	**0.82 / 0.64**	**0.86 / 0.71**	**0.40 / 0.34**
	Local Streets	0.25 / 0.23	**0.34 / 0.31**	**0.50 / 0.40**	0.20 / 0.18

In the MLR models and GAMs, the model general R^2s stand for the percentages of variance explained by the predictive variables. The results in Table 2.3 show that the GAMs explain higher percentage of variance than the MLR models with the same dataset. Compared with the GAMs, the MLR model CV R^2s are closer to the general R^2s, which means the MLR models are less over fit than the GAMs.

In addition to the AR(1) structure, the autoregressive-moving-average (ARMA) variance structure was also tested in sMLR models and sGAMs, but the models with ARMA structure didn’t perform better than the same models with AR(1) structure by comparing the CV results. In this study, the on-road data were collected on moving vehicles, and both the temporal and spatial variance need to be considered at the same time. This result shows that the ARMA structure doesn’t improve the AR(1) in reflecting the simultaneous temporal and spatial variance in the collected measurements.

The developed models fit arterial road data best. The possible reasons include a) the predictors work on the arterial road data better than freeway and local street data. For example, although the UFP levels in arterial roads were in the similar range with the UFP levels in local streets, but the vehicle speed range is greater in arterial road than in local street; b) The AADT data are only available for freeways and arterial roads, but not for local surface streets (Figure 2.1 and Table 2.1). This limited the performances of the local street models.

2.4.3 Selected variables

The predictive variables selected by the final models are summarized in Table 2.4. All models include most of the continuous meteorological parameters such as the on-road temperature / relative humidity or the ambient temperature / relative humidity, and the north-south wind speed (windNS) / east-west wind speed (windEW). In the MLR and sMLR models, the wind information (windNS and windEW) was dropped by the arterial road UFP models and arterial road $PM_{2.5}$ MLR model. The smooth function of windNS and WindEW was selected by all GAM and sGAM except for the arterial road sGAM (Table 2.4).

Season as a dichotomous meteorological predictor was dropped by all arterial road UFP models but selected by all local street $PM_{2.5}$ models. Also as a dichotomous traffic predictors, weekday was dropped by all freeway UFP models, but selected by all arterial road and local street UFP models, except for the local street sMLR model. In the $PM_{2.5}$ models, weekday was selected by all models except for the arterial road sGAM. As a continuous traffic predictor, AADT was selected by all freeway and arterial road UFP and $PM_{2.5}$ models, except for freeway $PM_{2.5}$ sGAM. Vehicle driving speed (speed or static) was selected by all UFP models except for the freeway sGAM. However, in $PM_{2.5}$ models, it was dropped by the freeway MLR and sMLR models, and the arterial road MLR model (Table 2.4).

As a spatial factor, altitude was selected by all local street UFP models but it was only selected by the freeway UFP sMLR model and the arterial road UFP GAM. Altitude was also selected by all $PM_{2.5}$ models except for arterial road GAM and sGAM. Land use was also selected by all local street UFP and $PM_{2.5}$ models, except for the local street $PM_{2.5}$ sGAM. However, it was dropped by almost all freeway and arterial road models. This suggests the spatial information, especially land use information, is more important in estimating the PM levels on local streets than those on arterial roads and freeways.

Table 2.4. Variable selection summary (* indicates $p < 0.05$)

		Models			
		MLR model	**sMLR model**	**GAM**	**sGAM**
UFP	**Freeway**	time* on-road temperature* windNS* speed* AADT*	s(time) on-road temperature*, humidity windNS* speed AADT altitude*	s(time) s(ambient temperature, humidity)* s(wnidNS, windEW)* season* s(speed)* s(AADT)*	s(time)* s(ambient temperature, humidity)* s(wnidNS, windEW)* season* s(AADT)* landuse*
	Arterial Road	**time*** **on-road temperature*,humidity*** **weekday*** **speed*** **AADT***	**s(time)*** **on-road temperature*, humidity*** **weekday*** **speed*** **AADT***	**s(time)*** **s(on-road temperature, humidity)** **s(windNS, windEW)*** **weekday*** **s(speed)*** **AADT** **altitude*, landuse***	**s(time)*** **s(ambient temperature, humidity)*** **weekday*** **s(speed)*** **AADT***
	Local Street	time* on-road temperature*, humidity* windNS* season* weekday* speed* altitude*, landuse*	s(time)* on-road temperature*, humidity* windNS* season* speed* altitude*, landuse*	s(time)* s(on-road temperature, humidity)* s(windNS, windEW)* season* weekday* s(speed)* altitude*, landuse*	s(time)* s(ambient temperature, humidity)* s(windNS, windEW)* weekday* s(speed)* altitude*, landuse*
$PM_{2.5}$	**Freeway**	time on-road temperature*humidity* windNS* season* weekday* AADT altitude*	s(time)* ambient temperature*, humidity* windNS season* weekday* AADT* altitude*	**s(time)*** **s(ambient temperature, humidity)*** **s(windNS, windEW)*** **season*** **weekday*** **s(AADT)*** **s(speed)*** **altitude***	s(time)* s(on-road temperature, humidity)* s(windNS, windEW) weekday s(speed)* altitude*
	Arterial Road	**time*** **ambient temperature*humidity*** **season*** **weekday*** **AADT*** **altitude***	**s(time)** **ambient temperature*, humidity*** **windNS** **weekday*** **static*** **AADT*** **altitude*landuse***	**s(time)*** **s(ambient temperature, humidity)*** **s(windNS, windEW)*** **weekday*** **s(speed)*** **s(AADT)***	**s(time)*** **s(ambient temperature, humidity)*** **s(windNS, windEW)*** **season** **s(speed)*** **s(AADT)***
	Local Street	time* on-road temperature*, humidity* windNS*, windEW* season* weekday* speed* altitude*, landuse*	s(time)* ambient temperature*, humidity* windNS season* weekday* speed* altitude, landuse*	**s(time)*** **s(on-road temperature, humidity)*** **s(windNS, windEW)*** **season*** **weekday*** **s(speed)*** **altitude*, landuse***	s(time)* s(on-road temperature, humidity)* s(windNS, windEW) season* weekday* s(speed)* altitude

2.4.4 Association and trends

The MLR models only describe the linear relationships between the predictors and the response variables, such as the on-road UFP and $PM_{2.5}$ in this study. The GAMs and sGAMs include both linear and non-linear relationships, and variable interactions. Comparing with the MLR models, the higher performance of GAMs indicate the importance of the non-linear relationships and the interactions of the predictors. However, the relationships and interactions usually become complicated when the degrees of freedom of the non-linear smooth functions increase.

The MLR and sMLR modeling results are summarized in Tables 2.5 and 2.6, respectively. Among the meteorological parameters, temperature and relative humidity were selected by almost all MLR and sMLR models, and their coefficients (slopes) were all positive, except for the on-road relative humidity in the freeway UFP sMLR model. These indicate positive associations between temperature / humidity and the on-road UFP / $PM_{2.5.}$ Specifically when the on-road or ambient temperature or relative humidity increase, so do the on-road UFP and $PM_{2.5,}$ while other variables in the model were held constant. Interestingly, windNS was dropped by all arterial road MLR and sMLR models except for the arterial road $PM_{2.5}$ sMLR model. When windNS was picked by the models, their coefficients were all positive. None of the MLR and sMLR models selected the windEW as a predictor for UFP and $PM_{2.5}$. These suggest that the north-south wind speed positively affects the on-road UFP and $PM_{2.5}$ concentrations, but the east-west wind doesn't affect the on-road UFP and $PM_{2.5}$. Similarly, the vehicle driving speed and AADT also affect the on-road UFP and $PM_{2.5}$ positively (Tables 2.5, 2.6). Weekday is a dichotomous variable, and the positive coefficients in the models indicate higher concentrations of UFP or PM$_{2.5}$ on regular weekdays and lower concentrations on weekends, while other factors are held constant. The modeling results showed that, the coefficients are consistently negative in PM$_{2.5}$ models, indicating the PM$_{2.5}$ levels on all road types were lower on regular weekdays than those on weekends, while other predictors are held same. The season slopes are all positive, showing that the warmer season levels were higher than the winter levels, while other predictors were constant.

Table 2.5. MLR model results (variable coefficient and SD listed)

	UFP			**PM$_{2.5}$**		
	Freeway	**Arterial Road**	**Local Street**	**Freeway**	**Arterial Road**	**Local Street**
Sample size (N)	1145	798	8277	840	608	8346
Intercept	**1.53×10^{1}** **(1.90×10^{-1})**	-2.17 (8.20×10^{-1})	**1.24×10^{1}** **(1.82×10^{-1})**	**2.93** **(4.73×10^{-1})**	6.44×10^{-1} (3.50×10^{-1})	**3.90** **(1.49×10^{-1})**
Predictive variables						
Time	**-2.59×10^{-5}** **(1.91×10^{-6})**	**-1.60×10^{-5}** **(4.29×10^{-6})**	**1.28×10^{-5}** **(8.45×10^{-7})**	**-2.67×10^{-5}** **(4.67×10^{-6})**	**8.76×10^{-6}** **(4.19×10^{-6})**	**-1.78×10^{-6}** **(6.70×10^{-7})**
Meteorological conditions						
On-road temperature	**8.67×10^{-3}** **(1.86×10^{-3})**	**1.38×10^{-1}** **(7.11×10^{-3})**	**1.47×10^{-2}** **(1.40×10^{-3})**	**3.08×10^{-2}** **(4.39×10^{-3})**	-	**1.15×10^{-2}** **(1.14×10^{-3})**
On-road relative humidity	-	**8.08×10^{-2}** **(4.86×10^{-3})**	**3.21×10^{-3}** **(1.02×10^{-3})**	**2.62×10^{-2}** **(3.81×10^{-3})**	-	**1.27×10^{-2}** **(8.25×10^{-4})**
Ambient temperature	-	-	-	-	**2.25×10^{-2}** **(4.19×10^{-3})**	-
Ambient relative humidity	-	-	-	-	**3.96×10^{-2}** **(1.44×10^{-3})**	-
Ambient wind NS	**3.15×10^{-2}** **(5.55×10^{-3})**	-	**4.14×10^{-2}** **(2.37×10^{-3})**	**2.09×10^{-2}** **(6.75×10^{-3})**	-	**1.36×10^{-2}** **(1.97×10^{-3})**
Ambient wind EW		-	-	-	-	-
Winter or summer		-	**5.83×10^{-2}** **(2.04×10^{-2})**	**1.79×10^{-1}** **(4.51×10^{-2})**	**3.37×10^{-1}** **(3.07×10^{-2})**	**4.28×10^{-1}** **(1.68×10^{-2})**
Traffic conditions						
AADT	**9.49×10^{-7}** **(2.79×10^{-7})**	**3.67×10^{-6}** **(4.52×10^{-7})**	NA	-4.03×10^{-7} (2.75×10^{-7})	**8.58×10^{-7}** **(2.04×10^{-7})**	NA
Speed	**1.28×10^{-3}** **(5.62×10^{-4})**	**1.31×10^{-2}** **(9.33×10^{-4})**	**6.12×10^{-3}** **(5.84×10^{-4})**	-	-	**2.17×10^{-3}** **(4.83×10^{-4})**
Static	-	-	-	-	-	-
Weekday or weekend	-	-	**9.97×10^{-2}** **(2.57×10^{-2})**	**-2.26×10^{-1}** **(8.29×10^{-2})**	**-1.53×10^{-1}** **(4.13×10^{-2})**	**-6.41×10^{-1}** **(2.06×10^{-2})**
Spatial features						
Land use						
Industrial	-	-	-4.44×10^{-2} (7.04×10^{-2})	-	-	2.36×10^{-2} (5.99×10^{-2})
Residential	-	-	**-1.87×10^{-1}** **(3.97×10^{-2})**	-	-	**-1.31×10^{-1}** **(3.52×10^{-2})**
Retail	-	-	**-1.67×10^{-1}** **(3.71×10^{-2})**	-	-	**2.49×10^{-1}** **(3.20×10^{-2})**
Others	-	-	**-1.48×10^{-1}** **(3.38×10^{-2})**	-	-	**-1.97×10^{-1}** **(2.95×10^{-2})**
Altitude	-	-	**-3.20×10^{-2}** **(1.39×10^{-4})**	**-4.20×10^{-3}** **(3.42×10^{-4})**	**-5.27×10^{-4}** **(2.12×10^{-4})**	**3.76×10^{-4}** **(1.13×10^{-4})**
Total variance explained	0.20	**0.55**	0.14	0.34	**0.82**	0.25
Cross validation R^2	0.17	**0.49**	0.10	0.22	**0.73**	0.23

Table 2.6. sMLR model results (degree of freedom, variable coefficient and SD listed)

		UFP			**$PM_{2.5}$**	
	Freeway	**Arterial Road**	**Local Street**	**Freeway**	**Arterial Road**	**Local Street**
Sample size (N)	1486	794	8290	874	616	8367
Intercept	**1.47 x 10^{1}** **(5.20 x 10^{-1})**	**-3.55** **(8.43 x 10^{-1})**	**1.23 x 10^{1}** **(1.66 x 10^{-1})**	**1.00** **(4.37 x 10^{-1})**	**1.63** **(4.35 x 10^{-1})**	**3.80** **(1.38 x 10^{-1})**
Predictive variables						
s(time) (d.f.)	**3.05** **(3.05)**	**8.66** **(8.66)**	**8.80** **(8.80)**	**7.87** **(7.87)**	**4.90** **(4.90)**	**8.81** **(8.81)**
Meteorological conditions						
On-road temperature	8.08 x 10^{-3} (4.90 x 10^{-3})	**1.43 x 10^{-1}** **(7.83 x 10^{-3})**	**2.27 x 10^{-2}** **(1.40 x 10^{-3})**	-	-	-
On-road relative humidity	-5.28 x 10^{-3} (2.95 x 10^{-3})	**8.62 x 10^{-2}** **(4.98 x 10^{-3})**	**3.97 x 10^{-3}** **(1.06 x 10^{-3})**	-	-	-
Ambient temperature	-	-	-	**3.91 x 10^{-2}** **(5.06 x 10^{-3})**	**1.64 x 10^{-2}** **(4.62 x 10^{-3})**	**5.83 x 10^{-3}** **(1.44 x 10^{-3})**
Ambient relative humidity	-	-	-	**3.16 x 10^{-2}** **(1.96 x 10^{-3})**	**4.13 x 10^{-2}** **(1.60 x 10^{-3})**	**1.77 x 10^{-2}** **(6.56 x 10^{-4})**
Ambient wind NS	**3.84 x 10^{-2}** **(6.87 x 10^{-3})**	-	**2.75 x 10^{-2}** **(2.40 x 10^{-3})**	**1.22 x 10^{-2}** **(6.45 x 10^{-3})**	**3.16 x 10^{-2}** **(4.60 x 10^{-3})**	**1.12 x 10^{-2}** **(1.77 x 10^{-3})**
Ambient wind EW	-	-	-	-	-	-
Winter or summer		-	**1.05 x 10^{-1}** **(2.02 x 10^{-2})**	**2.47 x 10^{-1}** **(4.94 x 10^{-2})**	-	**3.28 x 10^{-1}** **(1.62 x 10^{-2})**
Traffic conditions						
AADT	5.51 x 10^{-7} (2.92 x 10^{-7})	**4.01 x 10^{-6}** **(4.26 x 10^{-7})**	NA	**-5.06 x 10^{-7}** **(2.54 x 10^{-7})**	**8.60 x 10^{-7}** **(2.27x 10^{-7})**	NA
Speed	1.16 x 10^{-3} (6.95 x 10^{-4})	**1.17 x 10^{-2}** **(9.55 x 10^{-4})**	**5.91 x 10^{-3}** **(5.70 x 10^{-4})**	-	-	**4.08 x 10^{-3}** **(4.32 x 10^{-4})**
Static	-	-	-	-	**-1.45 x 10^{-1}** **(4.39 x 10^{-2})**	-
Weekday or weekend	-	-	-	**-3.60 x 10^{-1}** **(8.14 x 10^{-2})**	**-1.81 x 10^{-1}** **(4.42 x 10^{-2})**	**-6.27 x 10^{-1}** **(1.82 x 10^{-2})**
Spatial features ***Land use***						
Industrial	-	-	1.89x 10^{-2} (6.86 x 10^{-2})	-	**2.51 x 10^{-1}** **(1.01 x 10^{-1})**	**-1.62 x 10^{-1}** **(5.23 x 10^{-2})**
Residential	-	-	**-1.87 x 10^{-1}** **(3.89 x 10^{-2})**	-	2.41 x 10^{-2} (6.10 x 10^{-2})	-5.20 x 10^{-2} (3.26 x 10^{-2})
Retail	-	-	**-1.90 x 10^{-1}** **(3.66 x 10^{-2})**	-	**1.46 x 10^{-1}** **(6.19 x 10^{-2})**	**3.13 x 10^{-1}** **(3.02 x 10^{-2})**
Others	-	-	**-1.48 x 10^{-1}** **(3.32 x 10^{-2})**	-	8.70 x 10^{-2} (5.49 x 10^{-2})	**-1.65 x 10^{-1}** **(2.73 x 10^{-2})**
Altitude	**7.62 x 10^{-4}** **(3.85 x 10^{-4})**	-	**-2.97 x 10^{-3}** **(1.34 x 10^{-4})**	**-4.69 x 10^{-3}** **(3.55 x 10^{-4})**	**-6.73 x 10^{-4}** **(2.35 x 10^{-4})**	2.00 x 10^{-4} (1.04 x 10^{-4})
Total variance explained	0.15	**0.63**	0.19	0.44	**0.82**	**0.34**
Cross validation R^2	0.07	**0.50**	0.11	0.21	**0.64**	**0.31**

The GAM and sGAM results are summarized in Tables 2.7 and 2.8, respectively. The modeling results showed that, same as in the MLR and sMLR models, the coefficients of "weekday" are consistently negative in the $PM_{2.5}$ models. These indicate that the regular weekday $PM_{2.5}$ levels are lower than the weekend levels, while other predictors are held same. Similarly, the coefficients of "season" in the $PM_{2.5}$ models are all positive when it was selected, indicating the warmer season has higher on-road $PM_{2.5}$ levels compared with the colder season while other predictors held same. However, the regional $PM_{2.5}$ sampling results show that the warmer season had lower ambient $PM_{2.5}$ in Los Angeles [56]. As mentioned, the on-road $PM_{2.5}$ levels are also affected by the coagulation of nuclei mode particles and the tire/brake wear of the vehicles. Usually, the higher temperature and more dynamic meteorological conditions can enhance the tire/brake wear of the motor vehicles and particle dispersion.

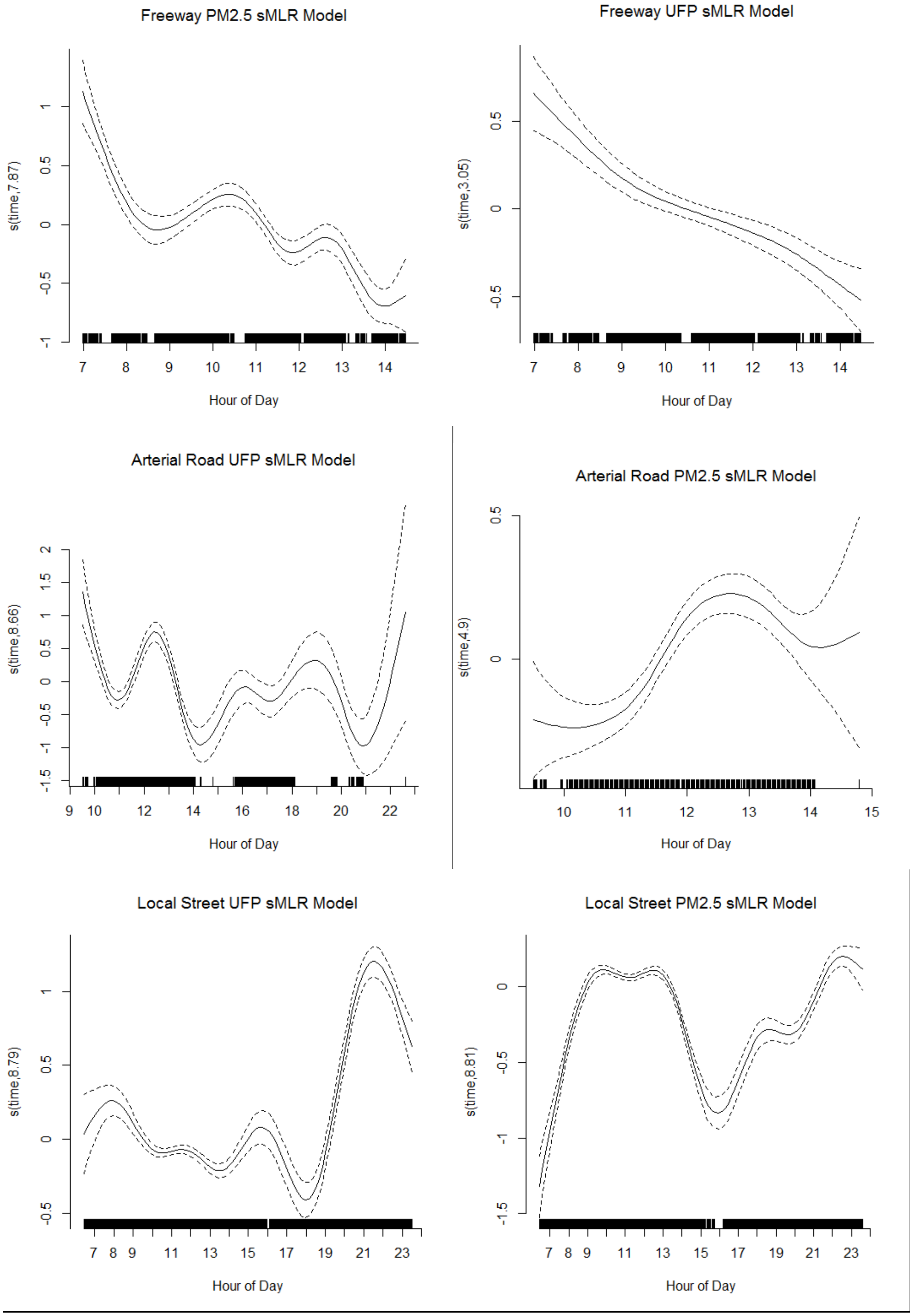
Table 2.7. GAM results (shaded area indicate the degrees of freedom of the smooth functions)

		UFP			$PM_{2.5}$	
	Freeway	**Arterial Road**	**Local Street**	**Freeway**	**Arterial Road**	**Local Street**
Sample size (N)	1515	582	8316	874	857	8367
Intercept	**1.53 x 10^{1}**	**1.34 x 10^{1}** **(1.80 x 10^{-1})**	**1.44 x 10^{1}** **(3.85 x 10^{-2})**	**5.65** **(7.32 x 10^{-2})**	**5.07** **(3.99 x 10^{-2})**	**5.29** **(2.87 x 10^{-2})**
Predictive variables						
f(time) (d.f.)	**5.28**	**4.14**	**8.34**	**4.86**	**4.92**	**7.71**
Meteorological conditions						
On-road temperature & relative humidity	**-**	**5.49**	**17.62**	**-**	**-**	**-**
Ambient temperature & relative humidity	**7.59**	**-**	**-**	**7.40**	**5.29**	**17.31**
windNS & windEW	**8.07**	**6.78**	**19.64**	**7.65**	**7.15**	**18.17**
Winter or summer	**-1.62 x 10^{-1}**	-	**4.92 x 10^{-2}** **(2.16 x 10^{-2})**	**2.25 x 10^{-1}** **(5.17 x 10^{-2})**	**-**	**3.43 x 10^{-1}** **(1.63 x 10^{-2})**
Traffic conditions						
AADT	-	7.24 x 10^{-7} (5.66 x 10^{-7})	NA	**-**	-	NA
f(AADT)	**4.33**	**-**	NA	**3.58**	**4.32**	NA
Speed	**-**	**-**	-	**-**	-	-
f(Speed)	**2.59**	**4.44**	**7.18**	**3.12**	**4.82**	**5.20**
Static	-	-	-	-	-	-
Weekday or weekend	**-**	**8.80 x 10^{-1}** **(1.55 x 10^{-1})**	**-7.10 x 10^{-2}** **(2.66 x 10^{-2})**	**-4.11 x 10^{-1}** **(7.86 x 10^{-2})**	**-3.25 x 10^{-1}** **(4.75 x 10^{-2})**	**-6.67 x 10^{-1}** **(1.78 x 10^{-2})**
Spatial features						
Land use						
Industrial	-	-1.64 x 10^{-1} (2.39 x 10^{-1})	-2.05 x 10^{-2} (6.53 x 10^{-2})	-	-	1.16 x 10^{-1} (4.64 x 10^{-2})
Residential	-	**-3.52 x 10^{-1}** **(1.45 x 10^{-1})**	**-1.90 x 10^{-1}** **(3.82 x 10^{-2})**	-	-	-3.06 x 10^{-2} (2.92 x 10^{-2})
Retail	-	**-4.59 x 10^{-1}** **(1.55 x 10^{-1})**	**-1.69 x 10^{-1}** **(3.64 x 10^{-2})**	**-**	-	**4.08 x 10^{-1}** **(2.77 x 10^{-2})**
Others	-	-2.69 x 10^{-2} (1.29 x 10^{-1})	**-1.47 x 10^{-1}** **(3.26 x 10^{-2})**	-	-	**-1.16 x 10^{-1}** **(2.45 x 10^{-2})**
Altitude	-	**-1.35 x 10^{-3}** **(5.50 x 10^{-4})**	**-2.87 x 10^{-3}** **(1.34 x 10^{-4})**	**4.05 x 10^{-3}** **(3.82 x 10^{-4})**	-	**4.89 x 10^{-4}** **(9.79 x 10^{-5})**
Total variance explained	0.26	**0.74**	0.28	0.50	**0.86**	**0.50**
Cross validation R^2	0.11	**0.54**	0.16	0.22	**0.71**	**0.40**

Table 2.8. sGAM results (shaded area indicate the degrees of freedom of the functions)

	UFP			**$PM_{2.5}$**		
	Freeway	**Arterial Road**	**Local Street**	**Freeway**	**Arterial Road**	**Local Street**
Sample size (N)	895	665	7669	662	712	7188
Intercept	**1.51x 10^{1}** **(7.72 x 10^{-2})**	**1.26x 10^{1}** **(2.74 x 10^{-1})**	**1.42x 10^{1}** **(4.71 x 10^{-2})**	**5.33** **(2.94 x 10^{-1})**	**4.84** **(1.63 x 10^{-1})**	**5.37** **(1.56 x 10^{-1})**
Predictive variables						
f(time) (d.f.)	**1.00**	1.00	1.00	**1.00**	**1.00**	2.39
Meteorological conditions						
On-road temperature & relative humidity	**-**		**-**	**-**	**-**	**24.77**
Ambient temperature & relative humidity	2.00	**2.00**	**13.10**	**2.00**	2.00	-
WindNS & WindEW	**3.03**	**-**	**11.36**	**2.00**	2.00	2.00
Winter or summer	**-5.51 x 10^{-2}**	-	-	-	2.73 x 10^{-1} (2.68 x 10^{-1})	**3.87 x 10^{-1}** **(1.28 x 10^{-1})**
Traffic conditions						
AADT	-	5.46 x 10^{-7} (6.68 x 10^{-7})	NA	-	-	NA
f(AADT)	**1.00**	-	NA	1.00	1.00	NA
Speed	-	-	-	-	-	-
f(Speed)	**-**	**2.84**	**3.07**	**3.84**	**3.25**	**1.00**
Static	-	-	-	-	-	-
Weekday or weekend	-	**1.30** **(2.97 x 10^{-1})**	-	-2.00 x 10^{-1} (3.27 x 10^{-1})	-	**-6.99 x 10^{-1}** **(1.62 x 10^{-1})**
Spatial features						
Land use						
Industrial	8.47 x 10^{-4} (5.54 x 10^{-2})	-	**1.14 x 10^{-1}** **(5.57 x 10^{-2})**	-	-	-
Residential	6.71 x 10^{-2} (5.68 x 10^{-2})	-	3.21 x 10^{-2} (3.69 x 10^{-2})	-	-	-
Retail	5.47 x 10^{-2} (7.62 x 10^{-2})	-	5.56 x 10^{-2} (3.71 x 10^{-2})	-	-	-
Others	**1.07 x 10^{-1}** **(5.16 x 10^{-2})**	-	2.78 x 10^{-2} (3.30 x 10^{-2})	-	-	-
Altitude	**-**	-	**-1.50 x 10^{-3}** **(3.72 x 10^{-4})**	**-1.71 x 10^{-3}** **(7.19 x 10^{-4})**	-	-1.55 x 10^{-4} (1.75 x 10^{-4})
Total variance explained	0.22	**0.49**	0.20	0.25	**0.40**	0.20
Cross validation R^2	0.15	**0.39**	0.18	0.22	**0.34**	0.18

In the sMLR models, time of day was set as the only non-linear predictor, and the resulted smooth functions were summarized in Figure 2.2. The time variable smooth functions in GAMs and sGAMs were summarized in Figures 2.3 and 2.4, respectively. The time trends show similarity in the sMLR models and GAMs, however, the time trends in sGAMs tend to be more linear and the degrees of freedom are close to 1. In most of the freeway and arterial road models, the time trends are negative, which indicates when the on-road UFP or PM2.5 levels go down when the time of day elapses. The time trends in local street models tend to be more complicated than them in arterial road and freeway models.



Freeway PM2.5 sMLR Model

Freeway UFP sMLR Model

Arterial Road UFP sMLR Model

Arterial Road PM2.5 sMLR Model

Local Street UFP sMLR Model

Local Street PM2.5 sMLR Model

Figure 2.2. Time variable smooth functions in sMLR models.

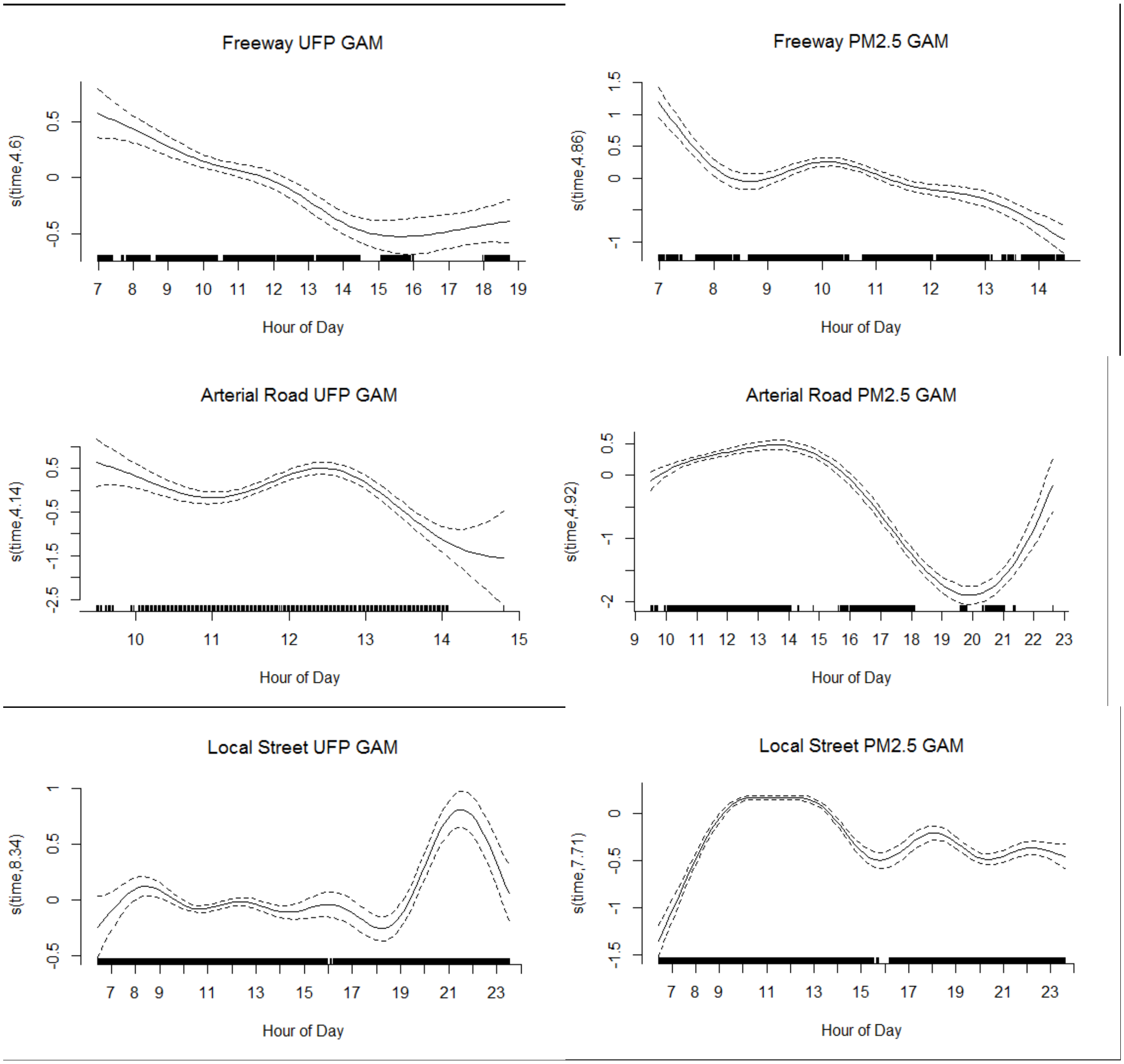


Figure 2.3. Time variable smooth functions in GAMs.

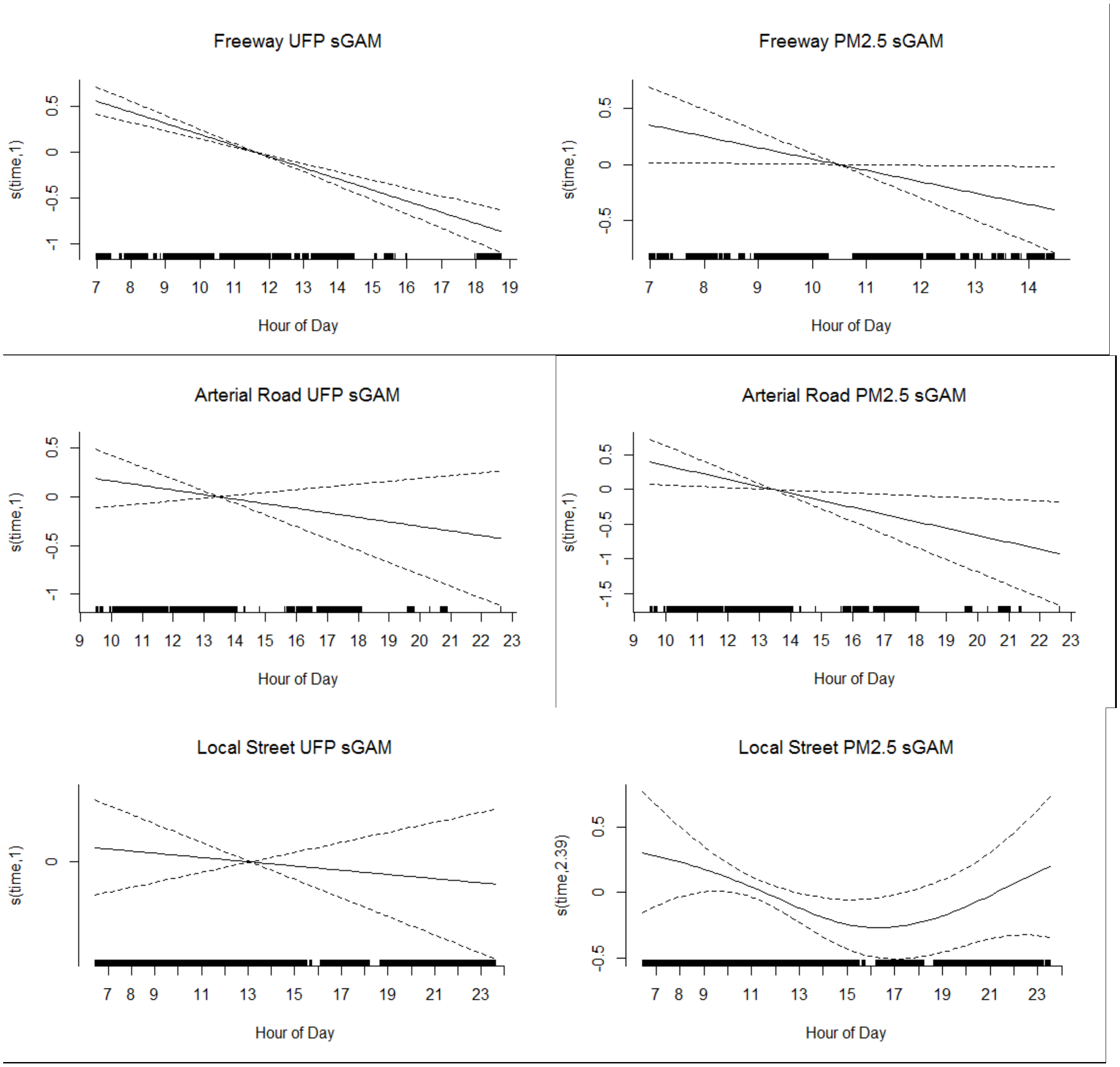


Figure 2.4. Time variable smooth functions in sGAMs.

In most of the GAMs and sGAMs, vehicle speed was a continuous predictive variable and fit in smooth functions with degrees of freedom higher than one. This makes the relationships between vehicle speed and UFP / $PM_{2.5}$ concentrations non-linear. The vehicle speed smooth functions in

GAMs and sGAMs are shown in Figures 2.5 and 2.6, respectively. The smooth functions show similar trends in the models with the same response variables, but the functions in GAMs usually have higher degrees of freedom than the smooth functions in sGAMs. When the vehicle driving speed goes up from 0, the smooth function results go up and reach their peak at 80-100 km/hr. When the speed was over 100 km/hr, the smooth functions go down slowly. The functions in the local street UFP models show higher degrees of freedom and the trends become more complicated (Figures 2.5 and 2.6).

The smooth functions of AADT in GAMs and sGAMs were shown in Figure 2.7. As Table 2.8 shows, the degrees of freedom of AADT smooth functions in sGAMs are all close to 1 (Table 2.8), but freeway UFP sGAM show positive trend, $PM_{2.5}$ sGAMs trends are almost flat (Figure 2.7). These indicate when the AADT goes up, the UFP levels go up when other predictors were held same. However, the AADT does not affect the on-road $PM_{2.5}$ levels in a very clear direction.

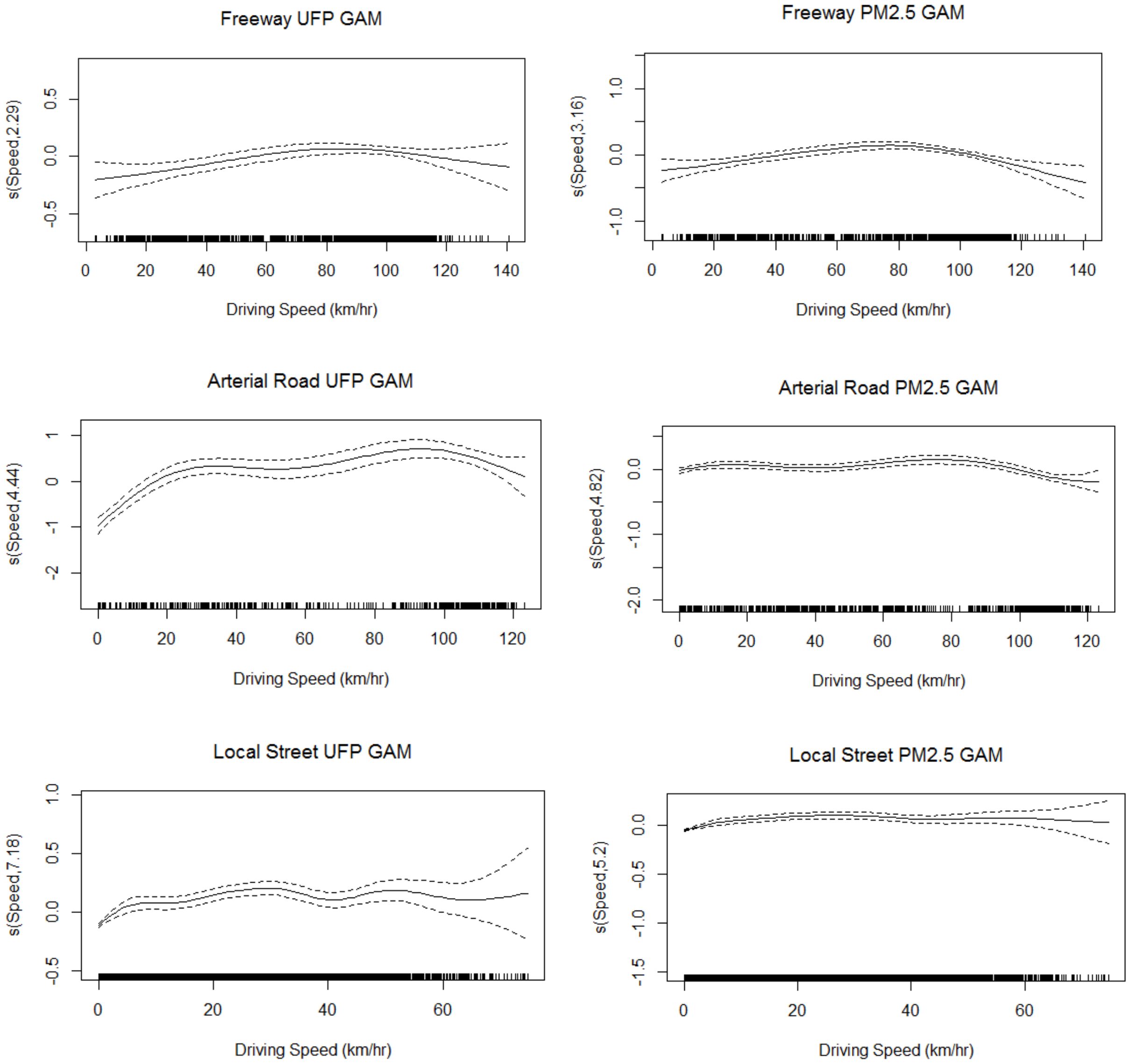


Figure 2.5. Smooth functions of vehicle speed in GAMs.

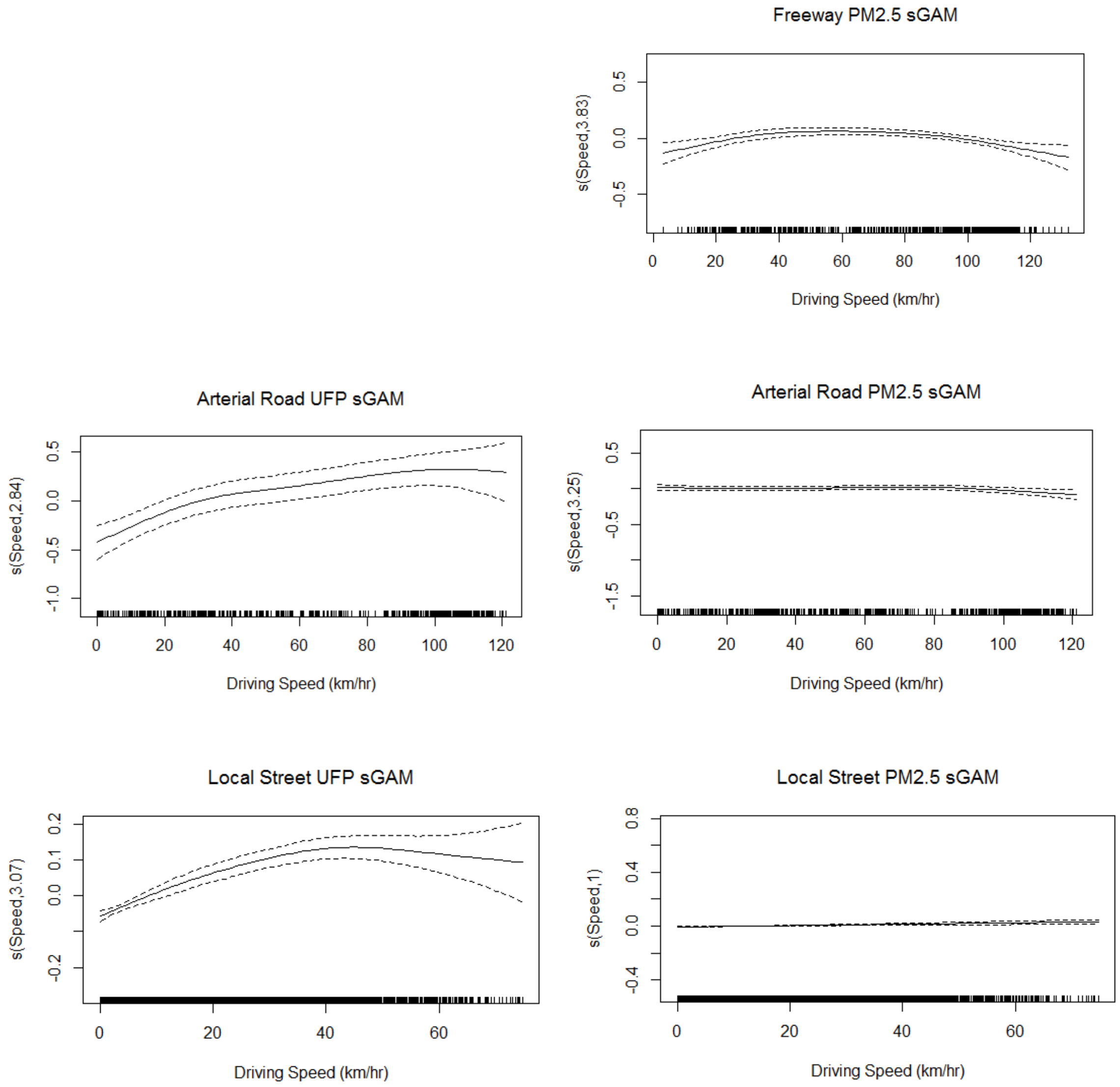


Figure 2.5. Smooth functions of vehicle speed in GAMs.

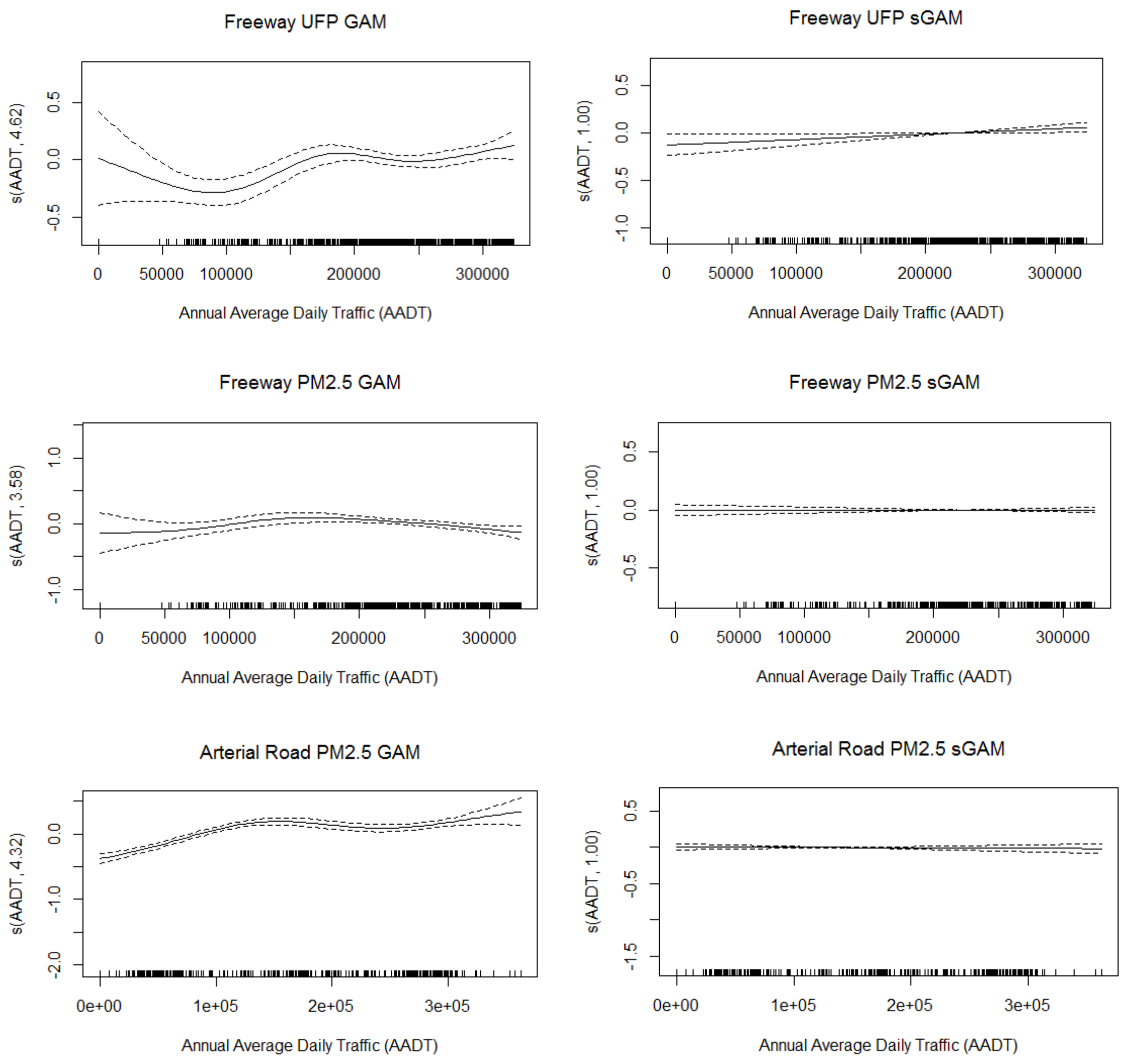
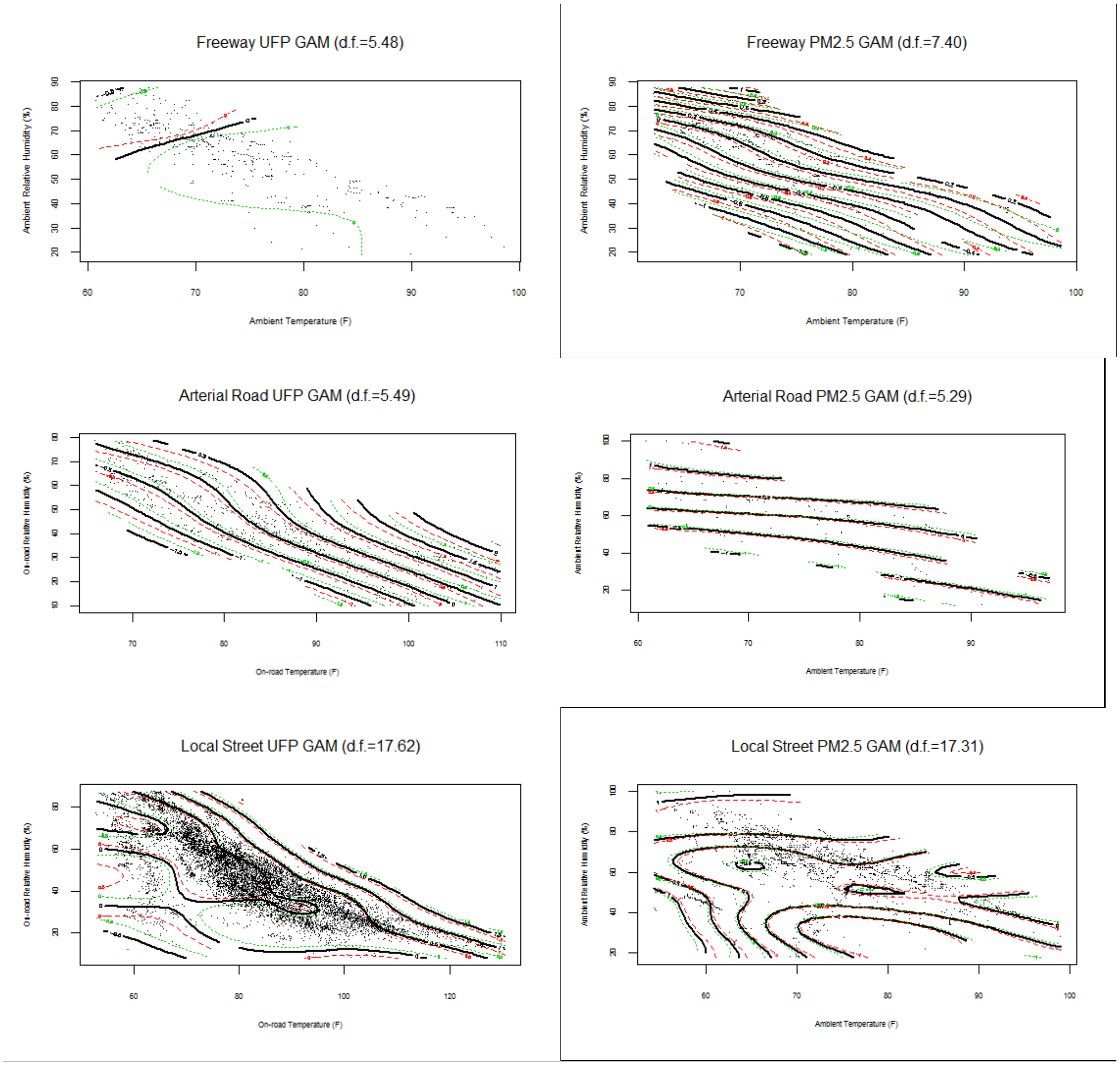


Figure 2.7. Smooth functions of annual average daily traffic (AADT) in GAMs and sGAMs.

The smooth functions of temperature and relative humidity in GAMs and sGAMs are summarized in Figures 2.8 and 2.9, respectively. In the GAMs, all the functions show that when the temperature and relative humidity go up, the function result go up, except for the functions in freeway UFP GAM and local street $PM_{2.5}$ GAM. In the freeway UFP GAM, when the temperature goes up and the relative humidity goes down, the UFP level goes up. In the local street $PM_{2.5}$ GAM, when the temperature goes down and the relative humidity goes down, the $PM_{2.5}$ level goes up. Similar as the time, speed and AADT smooth functions, the temperature and relative humidity functions in sGAMs have lower degrees of freedom compared with the GAMs, with the exception of local street $PM_{2.5}$ models. The degrees of freedom in the functions in freeway and arterial road sGAMs were all reduced to around 2.00. The trends in sGAMs are similar with the trends in most of the GAMs and linear models, that when the temperature and relative humidity go up, the UFP or $PM_{2.5}$ levels go up when other predictors held constant.

The smooth functions of windNS and windEW are summarized in Figures 2.10 and 2.11, respectively. In both GAMs and sGAMs, the wind functions show that when both the windNS and windEW are approaching 0, the UFP and $PM_{2.5}$ levels are going up in the freeways and local streets, but going down in the arterial roads, while other predictors were held same. Similar as the temperature and humidity functions, the smooth function degrees of freedom in the sGAMs were reduced from the degrees of freedoms in the GAMs. Previous studies show that the wind speed negatively affects both UFP and $PM_{2.5}$ concentrations on roadways, but the data collected on the arterial roads in this study show the contrary effects [9].


Freeway UFP GAM (d.f.=5.48)

Freeway PM2.5 GAM (d.f.=7.40)

Arterial Road UFP GAM (d.f.=5.49)

Arterial Road PM2.5 GAM (d.f.=5.29)

Local Street UFP GAM (d.f.=17.62)

Local Street PM2.5 GAM (d.f.=17.31)

Figure 2.8. Smooth functions of temperature and humidity in GAMs.

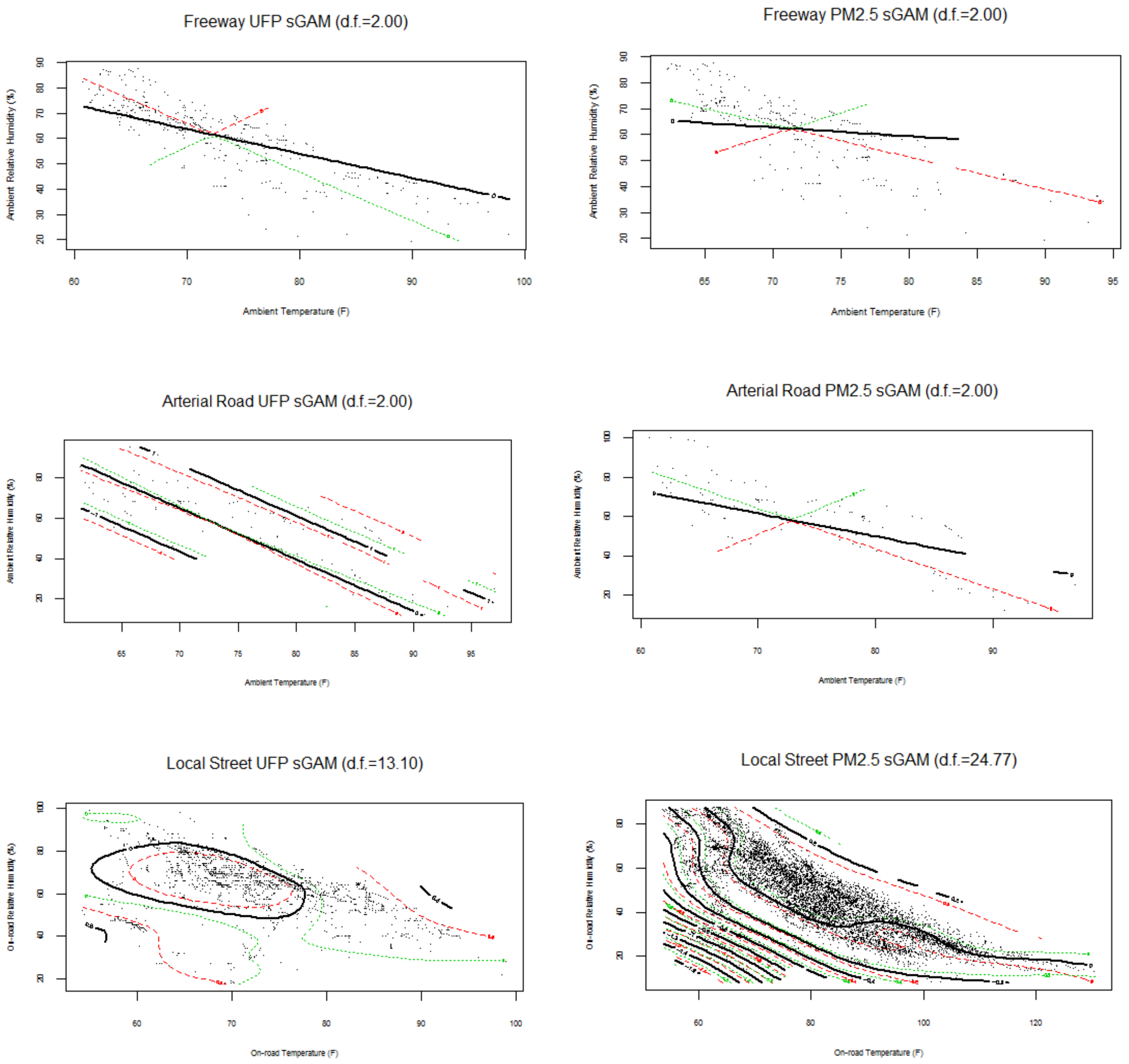


Figure 2.9. Smooth functions of temperature and relative humidity in sGAMs.

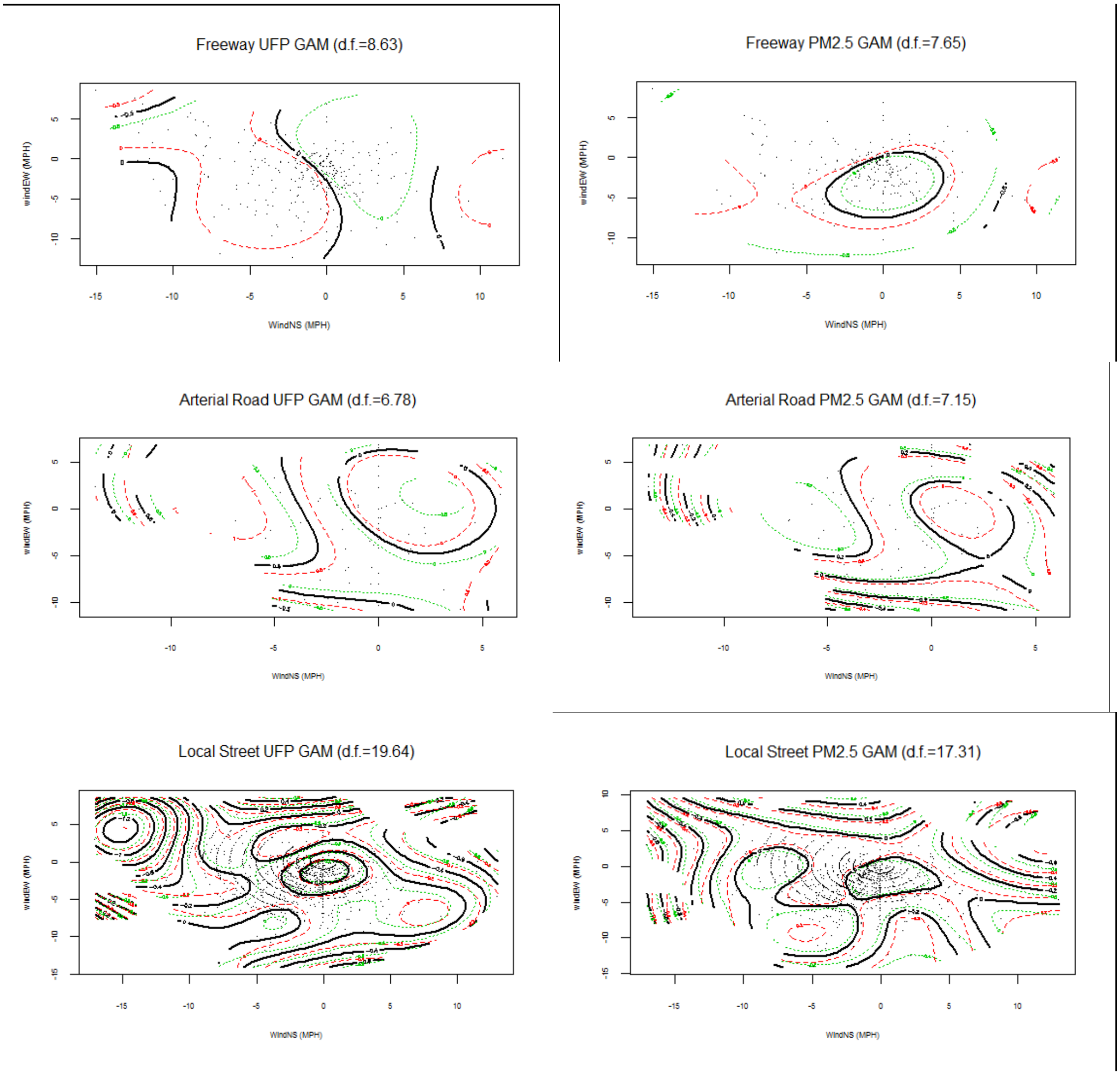


Figure 2.10. Smooth functions of windNS and windEW in GAMs.

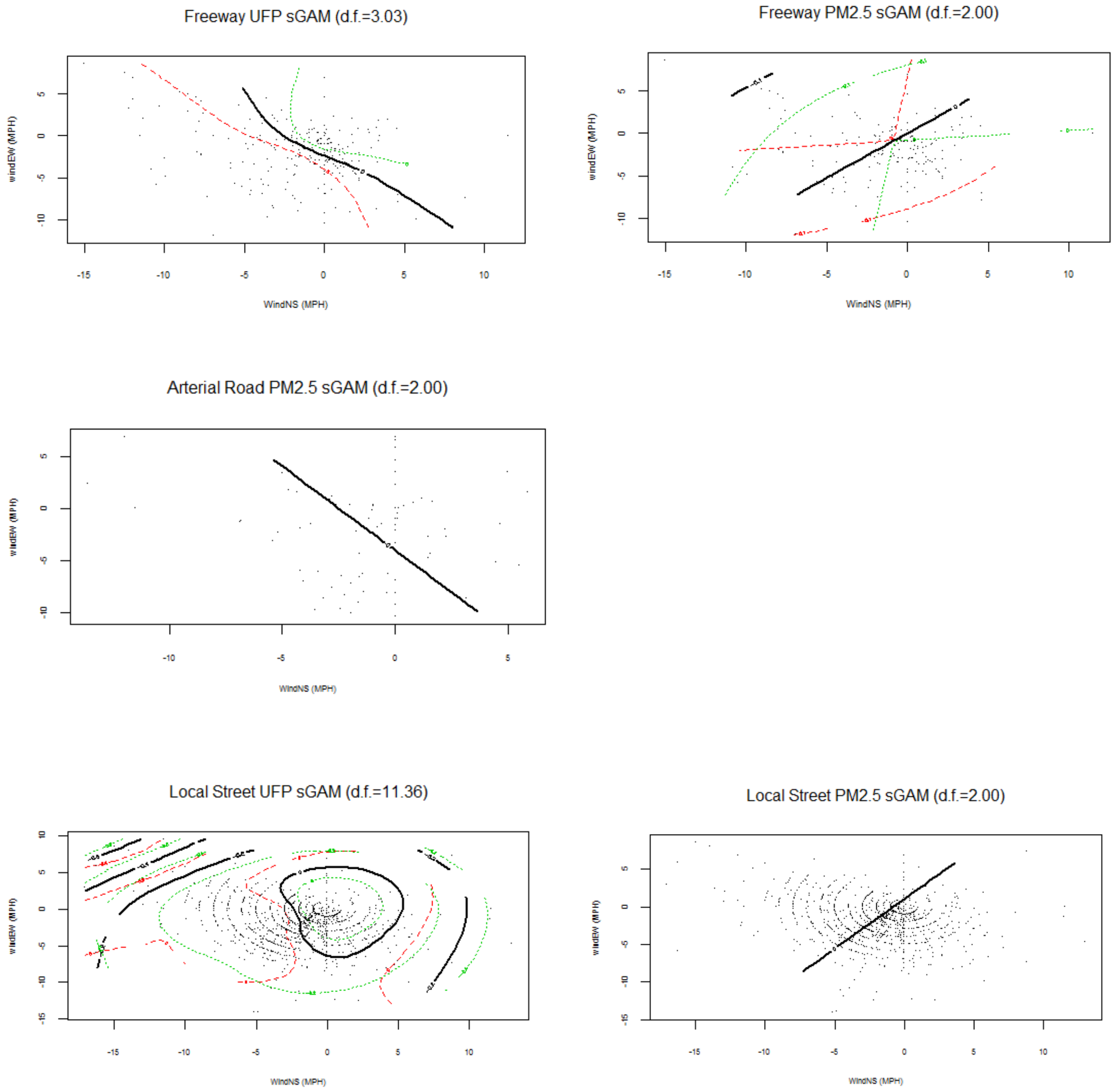


Figure 2.11. Smooth functions of windNS and wind EW in sGAMs.

3 TAXI IN-CABIN PARTICLE AND I/O RATIO

3.1 Abstract

Taxi drivers and passengers are exposed to high levels of fine ($PM_{2.5}$) and ultrafine particles (UFPs) but their exposures and related mitigation strategies are rarely explored. In this study, UFP and $PM_{2.5}$ concentrations were monitored concurrently inside and outside of 22 taxis under different ventilation and mitigation conditions. The results show that under realistic working (no mitigation; NM) condition, the average UFP and $PM_{2.5}$ levels inside taxis were 2.57×10^4 particles / cm^3 and 26 μg / m^3, and the average in-cabin to on-roadway (I/O) ratios for UFP and $PM_{2.5}$ were 0.60 and 0.75, respectively. When the taxi ventilation was set to outside air mode to avoid in-cabin carbon dioxide accumulation, and the windows were closed with the operation of high efficiency cabin air filter (WC+HECA), the UFP and $PM_{2.5}$ I/O ratios had the most substantial reduction to 0.47 and 0.52, respectively. Among the five tested taxi vehicle models, the Toyota Prius taxis exhibit the lowest UFP and $PM_{2.5}$ I/O ratios under both NM and the most stringent mitigation (WC+HECA) conditions. Compared to the originally equipped filter, the HECA filter also reduced the UFP and $PM_{2.5}$ I/O ratios most substantially in Toyota Prius taxis when the vehicle ventilation was set to outside air mode and the windows closed.

3.2 Introduction

High concentrations of particulate matter (PM), such as fine ($PM_{2.5}$, aerodynamic diameter ≤ 2.5 μm) and ultrafine particles (UFPs, diameter ≤ 100 nm), are usually observed on and near roadways [1-3]. PM has been shown to induce oxidative stress, mitochondrial damage, and acute pulmonary inflammation [31-33]. Previous studies have also reported an association between PM exposure and cardiorespiratory diseases as well as increased morbidity and mortality [57-59]. The International Agency for Research on Cancer (IARC) Working Group unanimously classified outdoor air pollution and PM as carcinogenic to human (IARC Group 1) [28].

Exposures to traffic that emit PM are of special concern for certain subpopulations, such as taxi drivers who spend more time in traffic than the general population. A report from the Los Angeles Department of Transportation showed that there were more than 4000 taxi drivers working in Los Angeles [13]. The exposures of taxi drivers to traffic emitted PM are likely to be much higher than people in other occupations, given their long work hours and the highly congested Los Angeles roadways [16, 17].

Previous studies have shown that in-cabin UFP levels are usually lower than on-road levels because of the different mechanisms of particle loss [11]. In fact, both on-road concentrations and the in-cabin to on-road (I/O) ratios determine in-cabin exposures. Research studies conducted on regular passenger vehicles with windows closed reported that for traffic related PM, the I/O ratios were 0-0.4 when the vehicle ventilation was under the in-cabin recirculation mode and were 0.6-1.0 under the outside air mode [10]. However, there is a knowledge gap on UFP and $PM_{2.5}$ levels inside taxis and related I/O ratios, which is essential for understanding the taxi vehicle protection against traffic related PM and estimating taxi driver and passenger exposures.

Previous studies have found that the significant determinants of UFP I/O ratios in passenger vehicles were ventilation modes, driving speed, cabin filtration performance, surface deposition and vehicle penetration [3, 11]. Other factors included vehicle model and car age/mileage [11, 12]. However, the way these factors may influence taxi vehicles, which have limited vehicle models but relatively higher age / mileage than regular passenger vehicles, is still poorly understood. Setting the vehicle ventilation to outside air mode and improving cabin filter efficiency is a promising mitigation strategy to reduce in-cabin UFP and $PM_{2.5}$ and avoid in-cabin carbon dioxide (CO_2) accumulation. Our previous studies showed that the high efficiency cabin air (HECA) filters reduced the in-cabin UFPs and $PM_{2.5}$ concentrations in passenger vehicles and school buses significantly under outside air mode when windows were kept closed [48, 49]. However, taxi vehicles were found to be more frequently used than passenger vehicles with higher air exchange rates (AERs) [12]. Thus, the effectiveness of these mitigation strategies in reducing PM inside taxis needs to be evaluated.

The hypothesis of this study is that the in-cabin PM levels are affected by the on-road levels, the mitigation strategies, and other factors, such as the car model, mileage, and driving speed. This chapter aims to (1) measure UFP and $PM_{2.5}$ levels concurrently inside and outside of taxis, (2) identify factors affecting the I/O ratios, and (3) evaluate the effectiveness of various exposure mitigation strategies for taxis.

3.3 Methods

3.3.1 Taxi recruitment

The detailed recruitment procedure has been described in a previous study [12]. Briefly, because taxi drivers earn at least twice as much on the one in five days that they are permitted to pick up fares at the Los Angeles International Airport (LAX), virtually every registered taxi driver in Los Angeles passes through the LAX taxi holding lot during that permitted day. The recruitment campaign was conducted at the LAX taxi holding lot for five consecutive days in March 2013, and a short recruitment form was designed and handed out to the taxi drivers. A total of 2449 blank forms were handed out, and 316 complete recruitment forms were collected, yielding a response rate of 13%. Out of these 316 taxi drivers, 121 who indicated never smoking cigarettes were eligible to participate in the study. Finally, 22 taxi drivers were selected randomly from the 121 eligible drivers stratified by their age and vehicle make/model. Information about their taxis is summarized in Table 3.1. The UCLA Institutional Review Board (IRB) approved the study design and experimental protocol in 2012. Informed consent was obtained from all participating taxi drivers.

Table 3.1. Summary of the Tested Taxi Models and Specifications

Taxi Type	**Maker**	**Model**	**N**	**Model Year**	**Average Mileage (mile) (SD)**	**Cabin Volume[1] (m^3)**
Hatchback	Toyota	Prius	10	2005-2012	114,000 (39,000)	3.3
Sedan	Ford	Crown Victoria	5	2005-2008	277,000 (93,000)	3.8
	Toyota	Camry	3	2009-2012	118,000 (46,000)	3.3
Minivan	Chevy	Uplander	1	2005	269,000	8.1
	Dodge	Grand Caravan	3	2007-2012	176,000 (82,000)	8.8

3.3.2 Ventilation and mitigation condition

The HECA filters were provided by an industrial partner, and installed into the taxi cabin filter holders by the researchers. These HECA filters were similar to originally equipped manufacturer (OEM) cabin air filters with respect to size, shape, and structure (i.e., the pleated panel type) but with different filtration media. The OEM filters were typically composed of a single layer of glass fibers, whereas the HECA filters were manufactured in double layer, with synthetic fibers on the upstream side and glass fibers on the downstream side, which achieved significantly higher filtration efficiencies than the OEM filters. The HECA filter manufacturer claimed that the filtration efficiency is equivalent to a minimum efficiency reporting value (MERV) rating of 16. The specifications and the scanning electron microscope (SEM) images of the HECA filter are reported elsewhere [48]. Because Ford Crown Victoria taxis do not have cabin filter holders, and portable high efficiency in-cabin air (HEPA) purifiers have been reported to be effective in reducing in-cabin particle levels [21, 60], a portable HEPA purifier (Model CWH3002, Honeywell Inc., Morris Plains, NJ) was used in the Ford Crown Victoria taxis to test mitigation effects. The air purifier had a built-in fan drawing air through a HEPA filter with a clean air delivery rate (CADR) of 25 m^3 / hour. During the sampling, the portable air purifier was placed in the rear of the cabin.

The experiments were designed to test various combinations of ventilation settings, window positions, and filter usage. In total, four conditions were tested for each taxi: (1) no mitigation (NM), to simulate realistic working conditions in which the ventilation/window was not controlled, and the HECA filter was not in use; (2) ventilation set to outside air to avoid CO_2 accumulation, and windows were closed, with the OEM filter or no filter in use (WC); (3) the ventilation was set to outside air to avoid CO_2 accumulation, and windows were closed with the HECA filter in use (WC+HECA), which is considered the most stringent mitigation strategy in this study; and (4) the ventilation and windows were not controlled, but the HECA filter was in use (HECA), which simulates realistic driving but uses the HECA filter as a simple engineering control. Every taxi was tested for four consecutive days under one of the four conditions on each day.

3.3.3 Driving routes

On each test day, the driver drove approximately 6 hours in the Greater Los Angeles area as he would typically do in his everyday work. A researcher rode along in the taxi to operate all the sampling instruments. On the first test day, each taxi driver was asked to drive from the start location (University of California Los Angeles) to the area where he usually worked and then repeat what he did in his previous work day. The testing routes included local streets, arterial roads, and freeways in the Greater Los Angeles Area. The start time of each test day and the driving routes were similar to minimize the differences in traffic and meteorological conditions for each individual taxi and driver. In total, field measurements were conducted on 83 different days from April to November 2013. Five test days were lost because two taxi drivers only partially completed their four-day tests.

3.3.4 Field measurements

In-cabin and on-road (outside taxi) $PM_{2.5}$ and UFP concentrations were concurrently monitored. Two identical sets of direct reading instruments were deployed for the measurements. One set monitored the in-cabin levels with tubing extended to the breathing zone of the driver. The other set monitored on-road concentrations with TSI isokinetic tubing extending outside through the back seat window. Three mm (id) tubing of the same length was used for both in-cabin and on-road sampling to ensure the same level of diffusion loss if any. The on-road air sampling conductive silicontubing was mounted onto the window. The window gaps were then sealed with heavy duty duct tape similar to the methods that were used previously for passenger vehicles [48].

Portable condensation particle counters (CPCs, Model 3007, TSI Inc., St. Paul, MN) were deployed to measure the UFP number concentrations. DustTrak monitors (Model 8520, TSI Inc., St. Paul, MN) with TSI $PM_{2.5}$ impactors installed at the inlets were used to measure $PM_{2.5}$ mass concentrations. The driving routes and speeds were recorded by a GPS unit (Qstarz GPS BT-1000XT, Taipei, Taiwan).

The in-cabin and ambient temperatures, relative humidity, carbon monoxide (CO) and CO_2 concentrations were measured simultaneously with two TSI Indoor Air Quality monitors (Q-Trak Plus, Model 8554, TSI Inc., St. Paul, MN). As both the in-cabin CO_2 emission rate due to human exhalation and the cabin volume are known, the AER can be estimated using CO_2 as a tracer gas. The detailed method for AER calculation and results have been presented in another study [12].

3.3.5 Data collection, analysis, and quality assurance

CPCs and DustTraks were zero calibrated prior to the field sampling and set to a logging interval of one second. DustTraks use a light scattering method to determine $PM_{2.5}$ mass concentration in real-time, which usually generates higher results than gravimetric methods. Hence the DustTraks were calibrated against simultaneous gravimetric measurements of $PM_{2.5}$ using a TEOM® (Series 1400A, Thermo Scientific Co., Waltham, MA), and a factor of 2.4 was achieved for data correction. The same correction factor was used in previous studies for $PM_{2.5}$ measurement correction [61, 62]. Each pair of instruments was collocated before and after the field sampling for data quality assurance. Good correlations with little bias were observed for both UFP and $PM_{2.5}$ with less than 5% of error and R^2 greater than 0.95.

The collected data were thoroughly checked, and unrealistic data points were removed, such as UFP readings that remained unchanged for more than a minute, which were usually due to low isopropanol levels or because the instrument was tilted. Negative and zero $PM_{2.5}$ and UFP readings generated when the built-in pumps were blocked were also removed. Approximately 85% of the collected data were used for further analysis. One-second raw data were averaged to five-minute data because PM measurements at the adjacent time points were highly correlated.

The I/O ratios were calculated from the five minute average in-cabin PM concentrations divided by the five minute average on-road concentrations. Other calculation methods such as using linear regression coefficients of in-cabin concentrations vs. on-road concentrations were explored but not used in this study, because the results are in the same range. SAS 9.4 software (SAS Institute, Cary, NC) was used for statistical analysis. *Student* t-tests or paired t-tests were used for two groups of data comparison. Analysis of variance (ANOVA) was used to compare multiple groups of data. The mixed effects linear regression method was used to evaluate variable associations. The model can be expressed by the following equation:

$$\log(I/O)_{ij} = \alpha + driver_i + \beta_1(vehicle\ mileage\ or\ age)_{ij} + \beta_2(mitigation)_{ij} + \beta_3(driving\ speed)_{ij} + \varepsilon_{ij}$$

Where i is the index of each driver, and j is the index of the calculated I/O ratio. α and β are the fixed intercept and slope respectively, and $driver_i$ is the random intercept of each driver. ε_{ij} is the residual.

Before t-tests, ANOVA and modeling, $PM_{2.5}$ and UFP concentrations and calculated I/O ratios were log transformed to achieve normal distributions (*Shapiro-Wilk* test $p > 0.05$). The level of significance was set as $p < 0.05$.

3.4 Results

3.4.1 Recruitment survey

Table 3.2 summarizes the taxi driver information collected from the 316 recruitment forms that were returned. It shows that Ford Crown Victoria (32.6%) and Toyota Prius (26.6%) were the most common models for taxi vehicles. Other vehicle models include Dodge Caravan (13.0%), Toyota Camry (7.8%), Chrysler Town & County (3.5%), Chevy Uplander (1.6%), and others (14.9%). The five vehicle models tested in this study (Toyota Prius, Ford Crown Victoria, Toyota Camry, Dodge Caravan, and Chevy Uplander) comprised 81.6% of all taxi vehicles in Los Angeles. Surveyed taxi model years ranged from 1987 to 2012, and 55.7% of the vehicles were more than 5 years old.

The survey also showed these taxi drivers have worked 11.9 (mean) ± 2.3 (SD) hours a day and 6.1 ± 0.8 days a week for 9.8 ± 8.3 years. They spent 4.2 ± 2.6 hours on freeways on each work day, and the taxi windows were open for 76.1 ± 12.4% of their driving time (Table 3.2).

3.4.2 Typical on-road and in-cabin PM concentrations

Since the UFP and $PM_{2.5}$ data were collected from moving taxis, the results reflect both temporal and spatial variations. Peaks were usually observed when the taxis were driving on freeways during traffic rush hours or adjacent to high emitters such as heavy duty trucks. Figure 3.1 shows the typical time series of simultaneously collected on-road and in-cabin UFP and $PM_{2.5}$ concentrations for a Toyota Prius taxi. The four tested conditions were marked on different sections in Figure 3.1. Rapid fluctuations were observed for both on-road UFP and $PM_{2.5}$ concentrations and the UFP levels changed faster and more substantially than $PM_{2.5}$. Comparing the patterns among different conditions, most of the concentration peaks occurred at a similar time of the day, which is mainly because the experiment was designed to have the similar start time and driving routes for each taxi driver to minimize the influences from traffic and meteorological changes. Although the on-road UFP and $PM_{2.5}$ concentrations showed similarity of patterns or trends, they were not necessarily at the same level among four test conditions. In fact, besides UFP and $PM_{2.5}$ on-road concentrations, UFP and $PM_{2.5}$ in-cabin concentrations were significantly different among the four mitigation conditions by ANOVA ($p < 0.05$).

Table 3.2. Summary of Los Angeles Taxi Driver Characteristics (n=316)

Category	Count (%)	Category	Count (%)
Age		BMI	
< 36	48 (15.2)	< 18.5	6 (1.9)
36-45	65 (20.6)	18.5-25	84 (26.6)
46-55	94 (29.7)	25-30	152 (48.1)
> 55	109 (34.5)	> 30	74 (23.4)
Ethnicity		Education	
Black	79 (25.0)	< high school	48 (15.2)
Hispanic/Latino	21 (6.6)	High school diploma or GED	55 (17.4)
White	128 (40.5)	Some college or associate's degree	81 (25.6)
Asian	58 (18.4)	Bachelor's degree	97 (30.7)
Others	30 (9.5)	Some graduate school or graduate	35 (11.1)
Marital status		Smoking status	
Married/Living with partner	225 (71.2)	Never	121 (38.3)
Divorced/Widowed	52 (16.5)	Quit	99 (31.3)
Never Married	39 (12.3)	Current smoker	96 (30.3)
Working out frequency		Self-reported health conditions	
0 days/week	133 (42.1)	heart conditions	18 (5.7)
1 day/week	76 (24.0)	hypertension	75 (23.7)
2 days/week	53 (16.8)	respiratory conditions	22 (7.0)
> 3 days/week	54 (17.1)	diabetes	38 (12.0)
Years as taxi driver			
<1	33 (10.4)	Workdays in each week	
1 - 5 years	74 (23)	< 5 days	10 (3)
5 - 10 years	70 (22)	5 days	34 (11)
10 - 20 years	91 (29)	6 days	177 (56)
>20	48 (15.2)	7 days	95 (30)
Driving time on freeways each work day		Open window time when driving	
<3 hours	112 (35)	<25% of time	50 (16)
3 - 6 hours	144 (46)	25 - 50%	89 (28)
6 - 9 hours	37 (12)	50 - 75 %	109 (34)
>9 hours	23 (7)	> 75%	68 (22)
Taxi vehicle make/model		Taxi vehicle age	
Ford Crown/Victoria	103 (32.6)	< 4	51 (16.1)
Toyota Prius	84 (26.6)	4 and 5	89 (28.2)
Dodge Caravan	41 (13.0)	6 and 7	91 (28.8)
Toyota Camry	25 (7.8)	8 to 10	68 (21.5)
Chrysler Towncounty	11 (3.5)	>10	17 (5.4)
Chevy Uplander	5 (1.6)		
Others	47 (14.9)		

The simultaneously collected on-road and in-cabin PM levels showed similar patterns as a function of time. As shown in Figures 3.1A and E, under the NM condition, when the taxi driver took short breaks, parked the taxi and opened the window at approximately 10:45-11:00 and 13:15-13:30, the in-cabin UFP concentrations were similar to outside levels. However, when the ventilation was set at recirculation mode for the rest of the day, the concentrations were lower than on-road levels. Similar effects of recirculation have been reported for passenger vehicles [3, 63]. Under the WC+HECA condition, UFP and $PM_{2.5}$ levels inside taxis were consistently lower than on-road levels (Figures 3.1C and 3.1G). Notably, under the WC condition, with the ventilation set to outside air mode, the in-cabin UFP levels were closer to on-road levels, which generated a higher average I/O ratio than that measured under the NM condition that was mainly in recirculation mode for this particular taxi (Figures 3.1A and 3.1B). Figures 3.1C and 3.1G show that under the WC+HECA condition and when the ventilation was set to outside air, the in-cabin UFP and $PM_{2.5}$ concentrations were still following the on-road trend, but the levels were much lower. This indicates that the HECA filter reduced both UFP and $PM_{2.5}$ levels inside the taxi and lowered their I/O ratios. Figures 3.1C and 3.1D, 3.1G and 3.1H show similar UFP and $PM_{2.5}$ I/O ratios between WC+HECA and HECA. The reason for this similarity is when testing WC and WC+HECA on the 2nd and 3rd test days, the driver was told to keep the windows closed and set the ventilation to outside air mode. Consequently, on the 4th test day (HECA condition), the driver tended to close the windows and set the ventilation to outside mode although he was allowed to control the ventilation and window position as he preferred. Figure 3.1 also shows missing data around noon when the driver turned off the engine and stepped out of the taxi for lunch breaks.

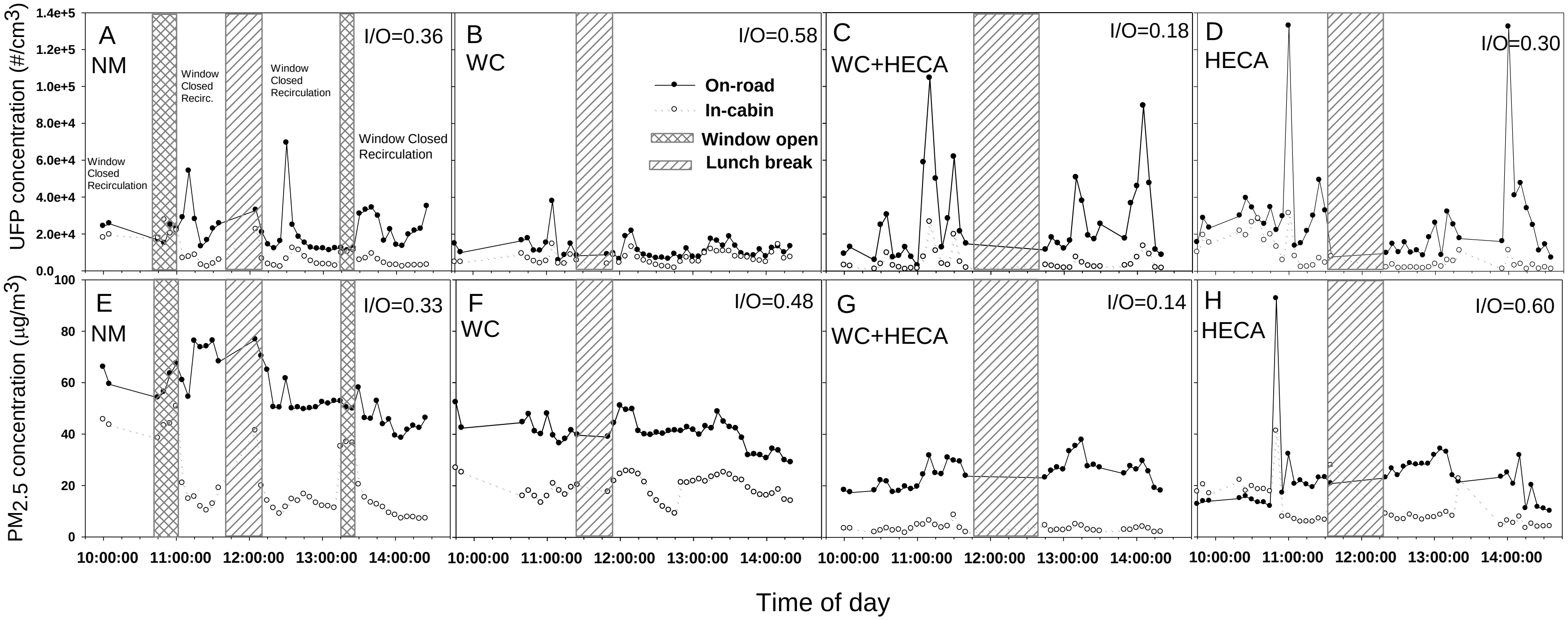


Figure. 3.1. Time series of UFP and $PM_{2.5}$ on-road and in-cabin concentrations under four test conditions for a Toyota Prius taxi. Solid lines show on-road (outside taxi cabin) concentrations and dotted lines show in-cabin concentrations. NM indicates “no mitigation”, WC indicates “window closed”, and HECA indicates “high efficiency cabin air filter in use”. The average I/O ratios under each condition were calculated and indicated on the figures.

3.4.3 Effects of ventilation and mitigation condition on PM levels and I/O ratios

Figure 3.2 summarizes the on-road and in-cabin UFP and $PM_{2.5}$ concentrations and their I/O ratios under four ventilation and mitigation conditions. It shows that throughout the study, the average on-road UFP concentrations ranged from 2.57×10^4 to 2.81×10^4 particles/cm^3 and the average on-road $PM_{2.5}$ concentrations ranged from 30 to 35 μg/m^3. The average in-cabin UFP and $PM_{2.5}$ concentrations under NM which simulates the realistic working condition without any mitigation, were 1.46×10^4 particles / cm^3 and 26 μg / m^3, respectively. These levels were approximately 40% and 25% lower than the on-road levels, with corresponding I/O ratios of 0.60 and 0.75.

When the ventilation was set to outside air mode under the WC+HECA condition, the average in-cabin UFP and $PM_{2.5}$ concentrations were approximately 53% and 48% lower than the on-road levels, with corresponding I/O ratios of 0.47 and 0.52, respectively. Compared with the other three conditions, WC+HECA generated the lowest I/O ratios.

Figures 3.2C and 3.2D summarized the calculated UFP and $PM_{2.5}$ I/O ratios under the four mitigation conditions. The mixed effects modeling results showed that UFP and $PM_{2.5}$ I/O ratios under the four different conditions were significantly different ($p < 0.05$). The lowest average UFP and $PM_{2.5}$ I/O ratios were found under the WC+HECA condition, and the highest average I/O ratios were found under the WC condition. This is because under NM, the ventilation was controlled by the driver. The ventilation was set to recirculation mode and the windows were kept closed for approximately 20 % of the NM testing time. This enabled the I/O ratios under NM to be lower than the I/O ratios under the WC condition, which has the ventilation always set to outside air mode.

Compared with NM, the average UFP I/O ratio decreased from 0.60 to 0.51, and the average $PM_{2.5}$ I/O ratio decreased from 0.75 to 0.61 at the HECA condition, where the ventilation and windows were not under control but the HECA filter was used. Compared with WC, the average UFP I/O ratio decreased from 0.65 to 0.47, and the average $PM_{2.5}$ I/O ratio decreased from 0.82 to 0.52 under the WC+HECA condition. These results indicate that when setting the ventilation to outside air mode and keeping the taxi window closed, the HECA filter reduced the UFP and $PM_{2.5}$ I/O ratios by 0.18 and 0.30, respectively, which doubled the I/O ratio reduction (0.09 for UFP and 0.14 for $PM_{2.5}$) when ventilation and window were not controlled.

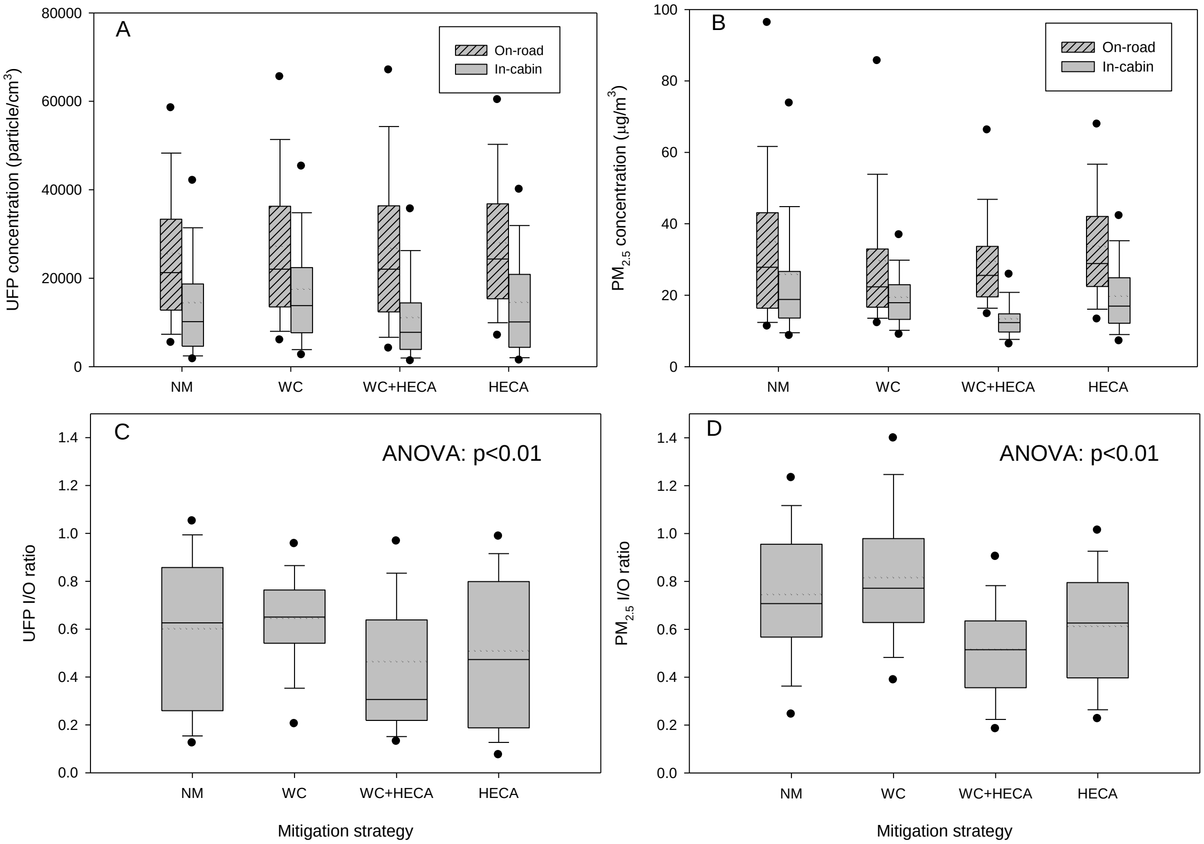


Figure 3.2. On-road and in-cabin UFP and $PM_{2.5}$ concentrations and corresponding I/O ratios under four different test conditions. The dotted line inside the box shows the mean, and the solid line inside the box show the median. The boundaries of the boxes are the quartiles of the data and the whiskers indicate the 10th and 90th percentiles. The dots represent the 5th and 95th percentiles. NM indicates “no mitigation”, WC indicates “window closed”, and HECA indicates “high efficiency cabin air filter in use”.

3.4.4 Other factors affecting I/O ratios

A previous study found that not only do ventilation settings affect taxi AERs but the car model, driving on freeway or local streets, driving speed, and car age/mileage can also play important roles [12]. These factors were investigated in this study for their effects on taxi I/O ratios.

Both UFP and $PM_{2.5}$ I/O ratios showed significant differences among the five tested taxi models by mixed effect modeling ($p < 0.01$). Figure 3.3 summarizes the taxi vehicle model effects on UFP and $PM_{2.5}$ I/O ratios. Under the NM condition, Toyota Prius had the lowest average UFP I/O ratio among all models, which shows that the Prius provides the best protection against on-road UFPs under realistic driving conditions. The Toyota Camry has the lowest average $PM_{2.5}$ I/O ratio under NM among the four taxi models (no data for Chevy Uplander due to instrument malfunction), which shows that the Camry provides the best protection against on-road $PM_{2.5}$ under realistic driving conditions.

Unlike other taxi models, the UFP I/O ratios for Toyota Prius taxis under WC (windows closed and ventilation set to outside air mode) increased when compared to NM (Figure 3.3). Under the WC condition, on average, the Toyota Prius taxis had the highest $PM_{2.5}$ I/O ratio and the second highest UFP I/O ratio among all tested vehicle models. The vehicle model with the highest average UFP I/O ratio under the WC condition is the Ford Crown Victoria, which do es not have an OEM cabin air filter. Thus, the Toyota Prius has the highest average UFP and $PM_{2.5}$ I/O ratios among all tested taxi models equipped with cabin air filters under the WC condition. In contrast, under both WC+HECA and HECA conditions, the Toyota Prius has the lowest UFP and $PM_{2.5}$ I/O ratios.

Unlike other taxi models, the I/O ratios of Ford Crown Victoria under the four conditions were not significantly different by ANOVA (Figure 3.3), suggesting that the mitigation strategies had limited effects on the Ford Crown Victoria. A possible reason is that the Ford Crown Victoria taxis were obtained from retired police cars with the highest car ages, mileage, and wear and tear. The average AER of the Ford Crown Victoria taxis was 75.2 h^{-1}, which is also much higher than the average AERs of the other four tested taxi models which range from 18.9 to 37.0 h^{-1} [12]. The higher AER in the Ford Crown Victoria rendered the portable air purifier less efficient than the HECA filters used in other taxi models.

As Table 3.3 shows, although the mixed effects linear regression results indicate no significant associations between I/O ratios and vehicle speed, UFP I/O ratios were significantly higher on freeways than on local surface streets, as determined by *Student* t-test ($p < 0.01$). Compared with levels measured on local streets, UFP and $PM_{2.5}$ concentrations were higher on freeways where the average vehicle speeds were also higher. This indicates that the UFP I/O ratios are sensitive to speed changes, but the association is not linear.

Table 3.3. Coefficients of mixed effect linear model for I/O ratios

	UFP I/O ratios			**PM2.5 I/O ratios**		
	Estimate	**Std. Err.**	**P-value**	**Estimate**	**Std. Err.**	**P-value**
Intercept	-1.19	0.17	<0.01	-0.54	0.15	<0.01
Mileage	3.06×10^{-6}	0.00	<0.01	9.58×10^{-7}	0.00	<0.01
Mitigation						
NM*	-	-	-	-	-	-
WC	0.07	0.04	0.08	0.06	0.03	0.02
WC+HECA	-0.36	0.04	<0.01	-0.38	0.03	<0.01
HECA	-0.19	0.04	<0.01	-0.15	0.03	<0.01
Speed	-1.40×10^{-4}	4.17×10^{-4}	0.74	1.44×10^{-4}	2.66×10^{-4}	0.59

*NM was used as the reference category.

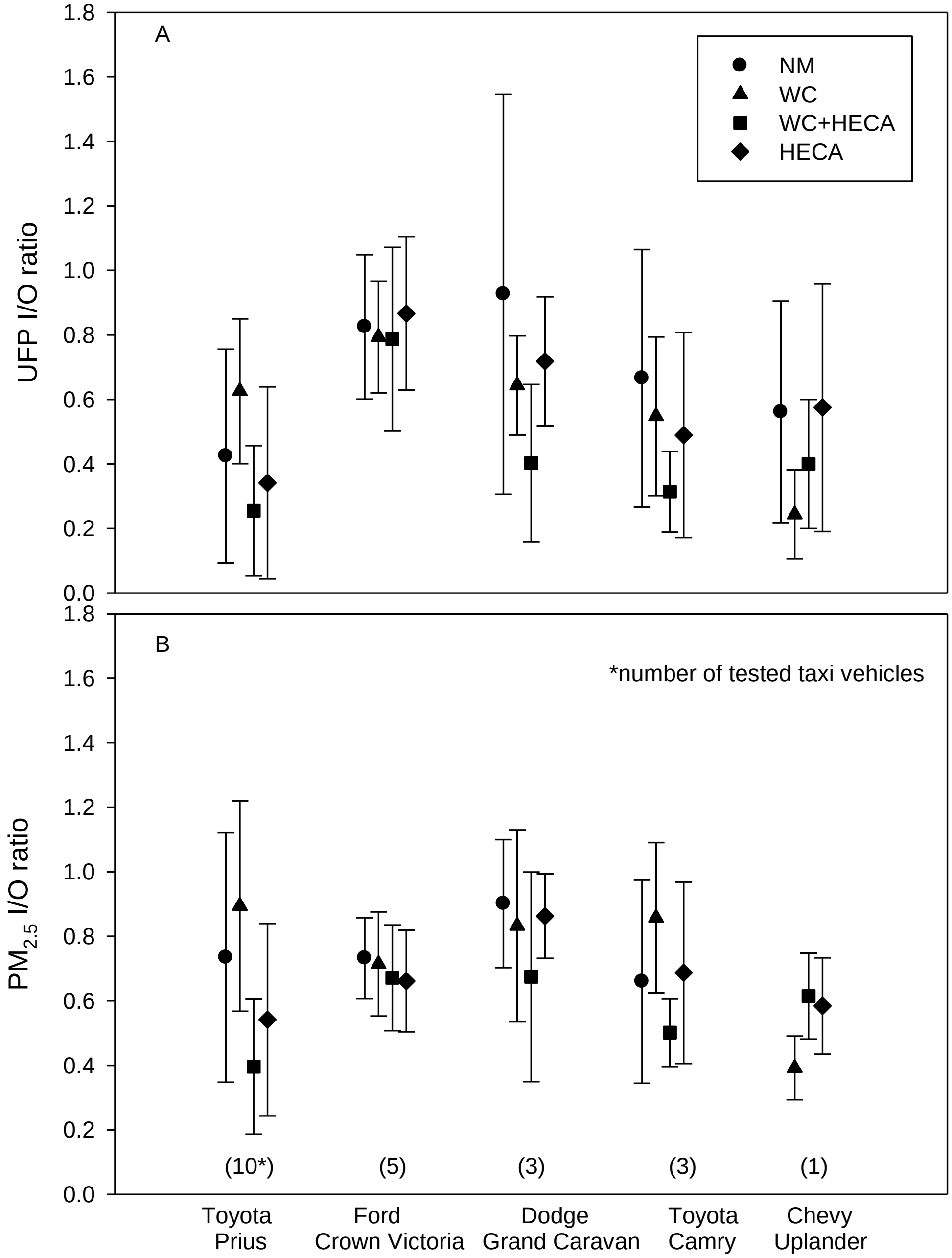


Figure 3.3. (a) UFP and (b) PM$_{2.5}$ I/O ratios for different taxi vehicle models. NM indicates "no mitigation", WC indicates "window closed", and HECA indicates "high efficiency cabin air filter in use".

3.5 Discussion

3.5.1. Comparison with passenger vehicles

Taxis typically have higher mileages than passenger vehicles of the same age. The taxis tested in this study had approximately twice as much mileage [12] as passenger vehicles of the same age reported in a previous study [64]. Therefore the tested taxis are expected to have more wear and tear and are likely to be leakier than regular passenger vehicles. It is reasonable to expect that the AER and I/O ratios of taxis are generally higher than those of regular passenger vehicles.

The mitigation strategies were found to be effective in reducing both UFP and $PM_{2.5}$ inside taxis, but the reduction rates were not as high as previously reported in passenger vehicles. Lee and Zhu reported that the HECA filters could reduce in-cabin UFP and $PM_{2.5}$ levels by 92% and 70% respectively [48]. Figure 3.4 compares the UFP and $PM_{2.5}$ sampling results of this taxi study with the study by Lee and Zhu on passenger vehicles [48]. When vehicles were tested using OEM filters, on-road UFP and $PM_{2.5}$ levels were all slightly higher in their study than those in the current taxi results, but the in-cabin UFP levels were lower in passenger vehicles (Figure 3.4). For vehicles tested using HECA filters, the on-road UFP and $PM_{2.5}$ levels in the study by Lee and Zhu were still higher, but the in-cabin levels were lower than in the current study. The calculated average I/O ratios in the passenger vehicles were always lower than the taxis regardless of which filter was in use. In addition, the UFP and $PM_{2.5}$ I/O reductions in passenger vehicles from using the OEM filter to using the HECA filter were over 60% higher than in taxis. This is mainly because the passenger vehicles tested in the study by Lee and Zhu were all less than three years old and more likely to be better maintained than the taxis. By comparison, 17 out of the 22 taxi vehicles tested in this study were older than three years, and about half of them were older than five years. Furthermore, the mileage on taxi vehicles was much higher, indicating they were more worn than regular vehicles of the same age [12, 64]. Thus, leaks from gaps around the HECA filter inside the ventilation system or from the cracks on doors, windows, and trunks may be higher in taxis than in the passenger vehicles.

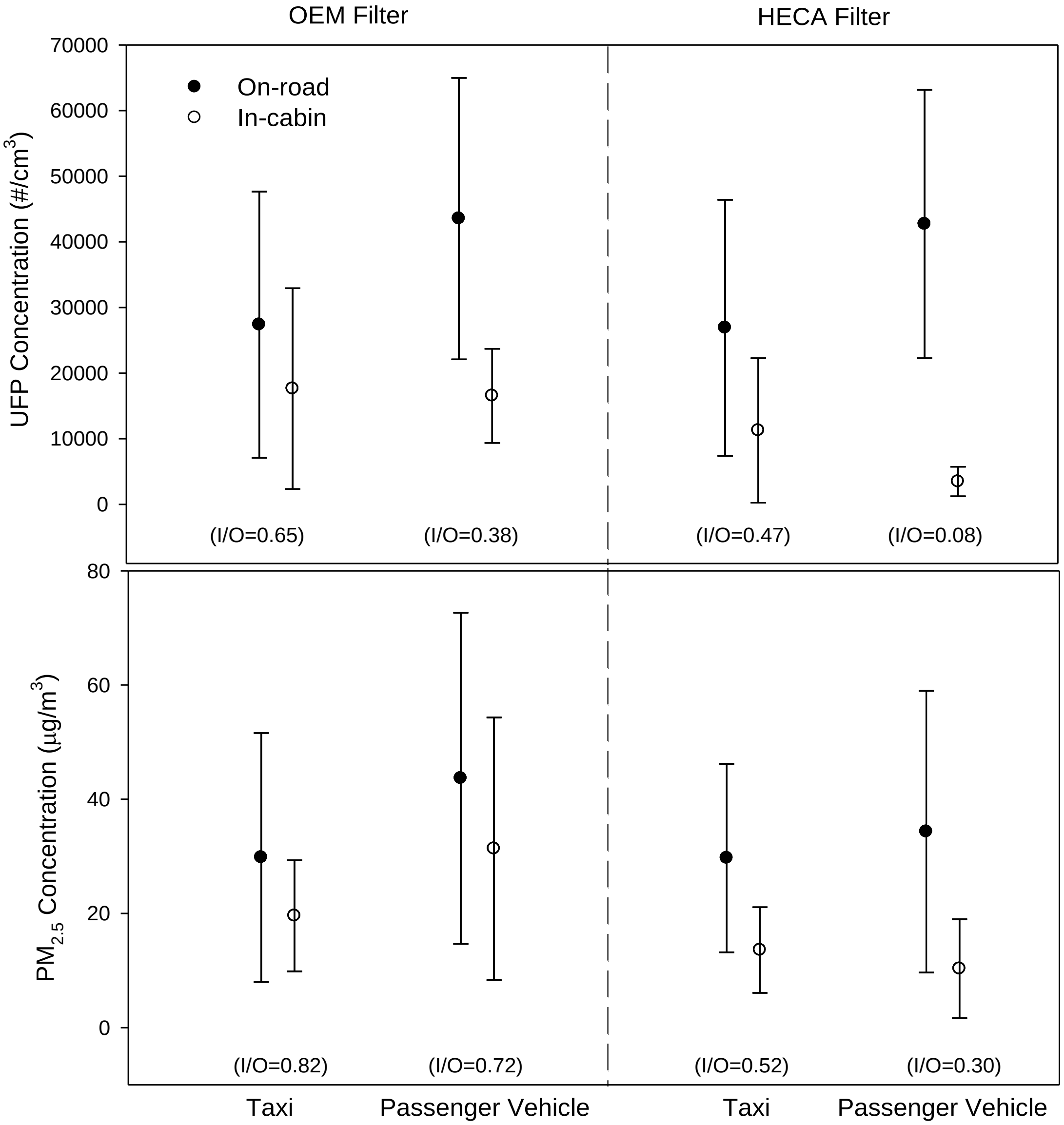


Figure 3.4. Comparison with passenger vehicles [48] for on-road and in-cabin UFP and $PM_{2.5}$ concentrations with OEM and HECA filters. The error bars indicate one standard deviation. Average I/O ratios were calculated and marked in parentheses.

Previous studies have showed that using a portable HEPA purifier could reduce UFP levels by 95-99% in passenger vehicles [21] and up to 50% of the total particles in school buses [60]. However, our results showed that the efficiency of air purifier was negligible in Ford Crown Victoria taxis. The portable air purifier used in this study operates at a CADR of 25 m^3/hour. The average AER of these Ford Crown Victoria taxis was 75.2 $hour^{-1}$, which was 3.3 times higher than the average AER of the other four tested taxi models [12]. The estimated volume of the Ford Crown Victoria taxis cabin is 3.8 m^3 (Table 3.1), and the total volume of air being exchanged in vehicle cabin was $75.2\ hr^{-1} \times 3.78\ m^3 = 284\ m^3 / hr$. This is approximately 11.4 times the CADR of the portable air purifier, rendering the effect of the air purifier negligible in removing the particles from the in-cabin air. The air purifier used in the study by Tartakovsky et al., had a higher CADR of 30 m^3/hour and lower vehicle cabin volumes that were between 2.0 and 2.5 m^3, which may help explain the higher efficiency observed in comparison to this study [21].

3.5.2 Effects of ventilation and mitigation condition and car model

The American Conference of Governmental Industrial Hygienists (ACGIH) has a 5000-ppm threshold limit value (TLV) for the 8-hour time weighted average (TWA) exposure of CO_2. The California Occupational Safety and Health Administration (Cal-OSHA) also has an 8- hour permissible exposure limit (PEL) of 5000-ppm CO_2. Exposure to CO_2 higher than 2500 ppm can significantly reduce decision making performances [65]. The in-cabin CO_2 accumulation is usually caused by the exhalation of the driver and passengers. Rapid in-cabin CO_2 accumulation has been observed in our previous studies when the vehicle ventilation was under the recirculation mode [3, 48]. In this study, the highest in-cabin CO_2 was observed under the NM condition, with an average concentration of 1220 ppm and ranging from 417 ppm to 4848 ppm. The maximum CO_2 level during NM (4848 ppm) was observed under the recirculation mode and is close to the occupational exposure limits of 5000 ppm. When the taxi ventilation was set to the outside air mode under WC and WC+HECA, the average CO_2 concentrations were 389 ppm and 408 ppm, respectively. Therefore, although the recirculation mode offers comparable or lower I/O ratios than the outside air mode with HECA+WC (Table 3.4), the CO_2 accumulation does not allow the recirculation mode to be a feasible in-cabin exposure mitigation strategy.

Among all the tested taxi models, the Toyota Prius was observed to have the lowest I/O ratios under both realistic (NM) and the ideal mitigation (WC+HECA) condition. However, the Toyota Prius had the highest I/O ratios among all taxi models equipped with cabin air filters when driven under the WC condition (Figure 3.3). This indicates Toyota Prius taxis provided the best protection against on-road UFP and $PM_{2.5}$ among the tested taxi models under both realistic working conditions and the ideal mitigation condition, but the least protection when windows were closed under outside air ventilation mode using OEM cabin air filters. The highest I/O under WC and the lowest I/O under WC+HECA suggested that switching cabin air filters reduced particle levels in Toyota Prius taxis more substantially than other taxi models.

Table 3.4. Summary of on-road and in-cabin PM concentrations and I/O ratios

Interventions	Mean (IQR[a])						ANOVA[b]
	NM[c]			WC[d]	WC[d]+HECA[e]	HECA	p-value
	Overall NM	***NM-RC+WC*[f]**	***NM-not RC+WC*[g]**				
$PM_{2.5}$							
On-road concentration (*$\mu g/m^3$*)	35 (16, 43)	*31* *(17, 42)*	*35* *(15, 40)*	30 (17, 33)	30 (20, 34)	34 (22, 42)	<0.01
In-cabin concentration (*$\mu g/m^3$*)	26 (14, 27)	*15* *(10, 18)*	*28* *(15, 27)*	20 (13.27, 22.93)	14 (9.72, 14.78)	20 (12, 25)	<0.01
Reduction[h] *(%)*	25 (5, 43)	*42* *(28, 63)*	*21* *(2, 37)*	34 (2, 37)	48 (37, 65)	39 (21, 60)	<0.01
I/O ratio[i]	0.75 (0.57, 0.95)	*0.58* *(0.37, 0.72)*	*0.79* *(0.63, 0.98)*	0.82 (0.63, 0.98)	0.52 (0.36, 0.63)	0.61 (0.40, 0.79)	<0.01
UFP							
On-road concentration *($x10^4$ particles/cm^3)*	2.57 (1.28, 3.34)	*2.44* *(1.25, 3.13)*	*2.61* *(1.27, 3.37)*	2.73 (1.35, 3.62)	2.69 (1.24, 3.64)	2.81 (1.53, 3.68)	0.09
In-cabin concentration *($x10^4$ particles/cm^3)*	1.46 (4.61, 1.87)	*0.73* *(2.37, 0.72)*	*1.72* *(6.64, 2.22)*	1.76 (7.67, 2.24)	1.13 (3.92, 1.44)	1.47 (4.37, 2.08)	<0.01
Reduction *(%)*	40 (14, 74)	*67* *(64, 87)*	*29* *(11, 50)*	35 (25, 56)	53 (36, 78)	49 (20, 81)	<0.01
I/O ratio	0.60 (0.26, 0.86)	*0.33* *(0.14, 0.36)*	*0.71* *(0.50, 0.89)*	0.65 (0.55, 0.76)	0.47 (0.22, 0.80)	0.51 (0.19, 0.80)	<0.01

a. IQR: interquartile range

b. ANOVA: analysis of variances. P-values reflected the significances of the differences among intervention groups

c. NM: no mitigation. taxi window and ventilation not under control

d. WC: taxi window all closed with outside air ventilation mode

e. HECA: high efficiency cabin air filter or high efficiency particulate air purifier in use

f. NM-RC+WC: no mitigation, taxi ventilation set to recirculation mode with window closed

g. NM-not RC+WC: no mitigation, but not under NM-RC+WC

h. Concentration reduction from on-road to in-cabin

i. In-cabin vs. on-road concentration ratio

Air filter collection efficiency (E) is defined as $\frac{C_{pre-filter}-C_{post-filter}}{C_{pre-filter}}$, where $C_{pre-filter}$ and $C_{post-filter}$ are the particle concentrations in air entering and leaving the filter, respectively. However, unlike the laboratory testing environment, the in-cabin particle concentration is not the same as the $C_{post-filter}$ because the outside air may enter the cabin through gaps and cracks. The particle deposition onto the vehicle interior surfaces leads to losses besides filter collection [66]. If it is assumed that the HECA filters in different taxi models had the same collection efficiency, the lowest I/O under WC+HECA indicates that Toyota Prius taxis were less leaky than others. It should be noted that the HECA filter efficiency in each model is also affected by other factors, such as the ventilation system design, the filter size/topology, and the instant pressure drop through the filter. Because of the limitation of this study, the collection efficiency of each filter used in taxi vehicles cannot be determined based on the collected data.

The highest I/O ratios under WC and the lowest under WC+HECA indicated that the HECA filter reduced UFP and $PM_{2.5}$ levels inside the Toyota Prius more significantly than other tested taxi models. There has been a trend of switching taxi vehicles to Toyota Prius because it is a hybrid vehicle with low emission and high mileage per gallon (MPG). The relatively light particle loads of the tested Toyota Prius taxi OEM cabin filters may be one of the reasons explaining their high I/Os under the WC condition. The results of this study suggest that improving the cabin filter efficiency will be an effective strategy to reduce both UFP and $PM_{2.5}$ levels inside the Toyota Prius.

3.5.3 Study Limitations

A total of 22 taxis were tested in this study, and thus, the number of each model was small. Some unpopular taxi models were not sufficiently sampled due to the stratification method. Because of the limited numbers of each model, linear regression was only applied to the Toyota Prius (n = 10) and Ford Crown Victoria (n = 5) to assess the taxis age/mileage effects. A positive correlation was found with I/O ratios for $PM_{2.5}$ ($p < 0.01$) but not for UFP. The ventilation settings and window positions were not recorded during the NM tests. Since the simultaneous rapid CO_2 build-up and UFP exponential decay indicate the vehicle uses the recirculation mode when the windows are closed [3], this condition was identified for all of the NM tests. However, other ventilation and window position combinations cannot be differentiated. The vehicle speed was calculated to 5 minute averages for correlation analysis with the 5-min average I/O ratios. Taxi speed changes rapidly in different roads and traffic conditions, and the recorded driving speed during this sampling campaign ranges from 0 to 133.9 km/hr, with an average of 34.6 km/hr and a standard deviation of 34.8 km/hr. The 5-min speed averages hardly reflect the rapid speed changes and are thus less likely to yield any associations with the measured I/O ratios. Finally, the day-to-day meteorology and traffic variations could not be fully controlled. In particular, although the ambient temperature fluctuated in a relatively low range in Los Angeles with recorded readings during the field sampling of 26.6 ± 6.9ºC (mean ± SD), it may have affected the decision to open windows when drivers were allowed to do so.

3.6 Conclusions

On average, Los Angeles taxi drivers are exposed to arithmetic means of 1.46×10^4 particles / cm^3 of UFP and 26 µg / m^3 of $PM_{2.5}$ while driving on the roadways without mitigation. The four ventilation and mitigation conditions of NM, WC, WC+HECA and HECA reduced the on-road UFP levels by 40%, 35%, 53%, and 4 %, and reduced on-road $PM_{2.5}$ levels by 25%, 34%, 48%, and 39%, respectively. UFP and $PM_{2.5}$ levels inside taxis, as well as their I/O ratios were significantly different under each of the four conditions. UFP and $PM_{2.5}$ had the lowest I/O ratios, 0.47 and 0.52, under the WC+HECA condition. Factors affecting I/O ratios and mitigation effects such as vehicle model, driving speed, and car age/mileage were analyzed. The results found that simply closing taxi windows (WC) but using outside air ventilation mode did not effectively reduce UFP and $PM_{2.5}$ I/O ratios in Toyota Prius taxis. The HECA filter reduced the PM in Toyota Prius taxis most substantially across all tested taxi models.

4 TAXI DRIVER PAH EXPOSURE AND LIPID PEROXIDATION

4.1 Abstract

Drivers or commuters who spend long time on roads are exposed to high levels of traffic related air pollutants (TRAPs) which have been associated with multiple adverse health effects. However, limited studies have investigated the effects of mitigation strategies in reducing the driver and commuters' exposures and their related health outcomes. In this study, the fine particulate matter ($PM_{2.5}$) and ultrafine particle (UFP) concentrations inside and outside 17 taxis were measured simultaneously while the taxis were driven on roadways. The drivers' urinary monohydroxylated polycyclic aromatic hydrocarbons (OH-PAHs) and malondialdehyde (MDA) concentrations just before and right after the driving tests were also determined. Data were collected under three driving conditions (i.e. no mitigation (NM), window closed (WC), and window closed plus using high efficiency cabin air filters (WC+HECA)) for each taxi and driver. The results show that, compared to NM, the WC+HECA reduced in-cabin $PM_{2.5}$ and UFP concentrations, by 37% and 47% respectively ($p < 0.05$), whereas the reductions on PAH exposures were insignificant. A statistically insignificant reduction of 17% was also observed in the drivers' urinary MDA under WC+HECA. The MDA concentrations were found to be significantly associated with the in-cabin $PM_{2.5}$ and UFP concentrations, suggesting the reduction of the drivers' lipid peroxidation can be at least partially attributed to the $PM_{2.5}$ and UFP reduction by WC+HECA. Overall, these results suggest using HECA filters has potential to reduce vehicle in-cabin particle (PM) levels and protect drivers' health.

4.2 Introduction

Air pollution is one of the leading risk factors of human premature death globally [18, 19]. As the most substantial combustion source in urban areas with dense population, vehicular emissions significantly increase carbon monoxide (CO), nitrogen oxides (NOx), fine particles ($PM_{2.5}$), ultrafine particles (UFPs), and polycyclic aromatic hydrocarbon (PAH) concentrations on / near roadways and inside vehicles [3, 20-22]. Over 600 million people worldwide are exposed to these hazardous traffic related air pollutants (TRAPs) [23, 24], which have been associated with various adverse health effects [25-27].

Among the TRAPs, $PM_{2.5,}$ UFP and PAHs have been found to be associated with cardiovascular diseases, which are the primary contributors to the air pollution related premature deaths [67]. Because of the frequently observed association between the exposures and the oxidative damage, the oxidative stress has been suggested to play a role linking the exposures with the cardiovascular diseases, although the mechanism is not fully understood. Specifically, previous studies have showed that $PM_{2.5}$ and UFP from traffic emissions have high oxidative potential that induces elevated systematic oxidative stress in rats and humans [68-70]. Malondialdehyde (MDA) is a lipid peroxidation end-product and usually produced under body systematic oxidative stress. Some *in vitro* and *in vivo* studies also suggest that PAHs and their metabolites can join the redox-active cycle and produce reactive oxygen species (ROS), which can attack larger biological molecules including polyunsaturated lipids and generate MDA [36, 37]. In addition, MDA has a potential to react with nucleic acid bases to form DNA adducts, create DNA interstrand, and even DNA protein cross-links to induce more health risks [40, 71].

Thus, MDA is a widely used biomarker for evaluation of oxidative stress and exposure related health effects [39, 40].

Vehicular emissions are known as the primary sources of ambient particulate matters (PM) in the Los Angeles air basin [72]. About 33-45% of total UFP exposure for Los Angeles residents occur due to time spent traveling in vehicles [2]. Some mitigation strategies have been explored in previous studies to reduce the commuter PM exposures, which include closing vehicle windows and using the high efficiency cabin air (HECA) filters. A passenger vehicle study found that keeping vehicle windows closed and using outside air ventilation mode reduced the on-road PM by up to 40% [73]. When the vehicle was equipped with a HECA filter, it reduced the on-road $PM_{2.5}$ and UFP by 70% and 92% respectively [74]. However, it remains unknown to what extent these mitigation strategies benefit commuters' health.

About 4,000 taxi drivers are working in the Greater Los Angeles area. They work 72 hours per week on average, and potentially have high TRAP exposures because of the long hours spent on roadways [14]. We hypothesize that the taxi driver exposures to UFP, $PM_{2.5}$ and PAHs will be reduced by the mitigation strategies, and the exposure reduction leads to peroxidation reduction. In this study the real time $PM_{2.5}$ and UFP concentrations inside and outside 17 Los Angeles taxis were measured while they were driven on roads, under different mitigation conditions. The drivers' urinary monohydroxylated-PAHs (OH-PAHs) and MDA concentrations were also measured just before and right after each monitored driving test. The aim of this study was to evaluate whether using HECA filters can effectively reduce the drivers' PM and PAH exposures, and the lipid peroxidation levels in their body.

4.3 Methods

4.3.1 Subjects

The detailed description on the taxi driver recruitment can be found elsewhere [12]. Briefly, in March 2013, 2449 recruitment forms were hand distributed to taxi drivers passing through the Los Angeles International Airport (LAX) taxi holding lot. A total of 316 completed forms were received back. Taxi drivers who smoked or drove Ford Crown Victoria taxis were excluded from this study, because cigarette smoking interferes with the PAH measurements, and the Ford Crown Victoria taxis were not equipped with a cabin air filter holder. Finally 17 were randomly selected out of the 90 qualified taxi drivers. Their demographic characteristics, body mass index (BMI), years as taxi drivers, and taxi vehicle make/models are summarized in Table 4.1. The study design and experimental protocol have been approved by the UCLA Institutional Review Board (IRB). Informed consents were obtained from all participants.

Table 4.1. Characteristics of the 17 studied taxi drivers.

	Mean ± SD	Minimum	Maximum
Age (year)	47 ± 13	28	67
Year as Taxi Driver	10 ± 6	2	20
BMI (kg / m^2)	26.8 ± 4.6	19.5	38.7
Gender, n (%)	16 males (94.1%)		
Ethnicity, n (%)		**Taxi Model, n (%)**	
Black	2 (11.8%)	*Chevy Uplander*	1 (5.9%)
Hispanic/Latino	0 (0%)	*Dodge Caravan*	3 (17.6%)
White	6 (35.3%)	*Toyota Prius*	10 (58.8%)
Asian	5 (29.4%)	*Toyota Camry*	3 (17.6%)
Other	4 (23.5%)		

4.3.2 Monitored driving test

Each driver was monitored for six hours a day and for four consecutive days, driving in the Greater Los Angeles area. The starting time and driving routes of each test day were kept consistent for each driver. During the tests the drivers were requested to mimic their actual work for driving, parking/waiting, and taking breaks during the monitored six hours.

On the first test day, the taxi drivers were allowed to control the window positions and use their originally equipped manufacturer (OEM) cabin filter to simulate their regular working conditions. On the second test day, the taxi drivers were requested to keep their vehicle windows closed, but their OEM cabin filter remained unchanged. The third test day is designed to have the most strict mitigation strategy in place by keeping the taxi windows closed and using a HECA filter simultaneously. The HECA filters were provided by an industrial partner and technical details of the filters can be found in Lee et. al 2014 [74]. Throughout the measurements, the taxi ventilation system was set to outdoor air mode to avoid CO_2 build-up. The fan was set to medium level when the windows were closed. On the fourth test day of each driver, the HECA filter was still in use, but the driver was controlling the ventilation and window position. However, from the observation, the drivers tended to close the vehicle windows on the fourth test day because of the request on the third test day, but their operation on the ventilation system was inconsistent. Thus the data from the fourth test days were not included in this study.

4.3.3 PM monitoring and urine collection

During the 6-hr monitored driving tests, portable condensation particle counter (CPC) (Model 3007, TSI Inc., St. Paul, MN) and DustTrak (Model 8520, TSI Inc., St. Paul, MN) were deployed inside taxi cabs to measure UFP and $PM_{2.5}$ concentrations, respectively. The inlets of two pieces of instrument were connected with the same length of TSI isokinetic tubing extending to the driver's breathing zone. Both CPC and DustTrak are real-time direct reading instrument with data logging function. UFP and $PM_{2.5}$ concentrations were logged with 1-sec interval during the experiments. The 6-hr averages were used for subsequent analysis. Urine samples were collected in 90 ml sterile screw cap urine containers just before and right after the 6-hr monitored driving tests (pre- and post-test). All collected urine samples were stored in a -20°C freezer until analysis.

4.3.4 Analysis of urinary OH-PAHs and MDA

The OH-PAH analysis was done in the California Department of Public Health (CDPH) laboratory. The method was based on enzymatic deconjugation, liquid-liquid extraction, trimethylsilylation of the OH-PAHs, followed by gas chromatography/isotope dilution high-resolution mass spectrometry [75]. Nine OH-PAHs were quantified (i.e. 1- and 2-hydroxynaphthalene (1- and 2-OH-NAP), 2-, 3- and 9-hydroxyfluorene (2-, 3-, and 9-OH-FLU), 1-, 2-, and 3-hydroxyphenanthrene (1-, 2-, and 3-OH-PHE), and 1-hydroxypyrene (1-OH-PYR)). The urinary MDA analysis was done in the State Key Laboratory of Peking University School of Environmental Sciences and Engineering. The method was explained in a previous study [41]. Briefly the MDA was analyzed by a high performance liquid chromatography (HPLC) system with a fluorescent detector. Each urine sample was added with a mixture of phosphoric acid and thiobarbituric acid (TBA, 42 mM), and incubated for 1 hour at 80 °C. Then the solution with MDA-TBA derivatives was injected into the HPLC system with a fluorescence detector at 532 nm. Urinary creatinine was analyzed with Jaffe method which generates stable creatinine results for urine samples kept at a temperature of -20ºC and thawed only once [76, 77]. The urinary OH-PAH and MDA concentrations were normalized to creatinine.

4.3.5 Quality assurance and quality control

For both CPC and DustTrak, flow rate measurement and zero check with HEPA filters was performed before and after each test. The DustTrak monitor was calibrated against simultaneous gravimetric measurements of $PM_{2.5}$. A factor of 2.4 was achieved and used for DustTrak data correction, which was consistent with data reported in previous studies [61, 62]. For OH-PAH analysis, quality control (QC) samples were analyzed along with the taxi driver samples to assure the precision and accuracy of the data. QC data were reviewed based on laboratory rules of quality control [78, 79]. Proficiency Test (PT) or inter-laboratory QC samples of unknown concentrations were performed twice a year for demonstration of OH-PAH analytical method performance.

4.3.6 Estimation of PAH exposure due to traffic

Urinary OH-PAHs are biomarkers of integrated body PAH intake through different exposure routes. The half-lives of the urinary OH-PAHs are usually several hours in human body, and it takes days to completely excrete OH-PAHs after the PAH exposures. Although the drivers tried to minimize their PAH intake from other sources by not eating barbequed or fried food during the test days, their urinary OH-PAH levels could still be influenced by the PAH exposures other than the traffic emissions during the monitored driving tests, such as previous TRAP exposures or dietary intake. Therefore, neither the pre- nor the post- test urinary OH-PAHs can directly indicate the traffic PAH exposure levels during the monitored taxi driver tests. Instead, a well-established one-compartment pharmacokinetic model with continuous infusion was used to estimate the traffic PAH exposure during the monitored tests [80]. In this model, constant and continuous exposure to PAHs during the tested six hours was assumed. In addition, it was assumed that PAHs undergo a first-order elimination in human body, which is widely supported by previous studies [81]. The average elimination rates of different PAH species were obtained from a previous study [82]. The calculated OH-PAH increments, denoted as $\text{OH-PAH}_{\text{trap}}$, were used as the surrogates of the PAH exposure during the driving tests.

The concentration of post-test OH-PAHs is expected to depend on both PAH exposure during the 6-hr monitored test and the baseline level of OH-PAHs (i.e. pre-test OH-PAHs concentration).

Since both pre- and post-test OH-PAHs levels were measured in this study, a pharmacokinetic model can be used to calculate PAH exposure during the 6-hour monitored test.

Previous studies indicated that elimination rate constants for urinary OH-PAH were first-order [82]. Assuming the urinary OH-PAH increment rate due to the continuous 6-hr TRAP exposure is E_i, the alteration of OH-PAHs in urine could be described as:

$$dc_i/dt = E_i - kC_i \tag{4.1}$$

Where i is an index for each driver, C_i is the concentration of OH-PAHs in urine, and k is the urine elimination rate of OH-PAHs in human body in hr^{-1}. Integrate equation (4.1) and get:

$$C_i(t) = C_i(0)e^{-kt} + E_i(1 - e^{-kt})/k \tag{4.2}$$

where $C_i(0)$ and $C_i(t)$ are the OH-PAH concentrations for driver i at initial condition (t=0) and time t, respectively. Figure 4.1 illustrates the two terms of equation (4.2) and the combination, where $C_i(0)\, e^{-kt}$ indicates the first order decay of the OH-PAH concentration from the initial condition, and $E_i\,(1 - e^{-kt}) / k$ indicates the OH-PAH concentration increment due to the continuous TRAP exposure during the 6-hr monitored test.

If the pre-test urinary concentration is set as the initial condition, the urinary OH-PAH increment at t = 6 hour due to the TRAP exposure ($C_{i,trap}$) can be calculated with the following equation:

$$C_{i,trap} = E_i(1 - e^{-kt})/k = C_{i,post} - C_{i,pre}e^{-6k} \tag{4.3}$$

In this equation, $C_{i,pre}$ and $C_{i,post}$ were measured, the value of k was obtained from previous studies for each OH-PAH species [82]. Thus, the increment of each and total OH-PAH (OH-PAH_{trap} and $\sum$OH-PAH_{trap}) due to the 6-hr monitored TRAP exposure can be calculated.

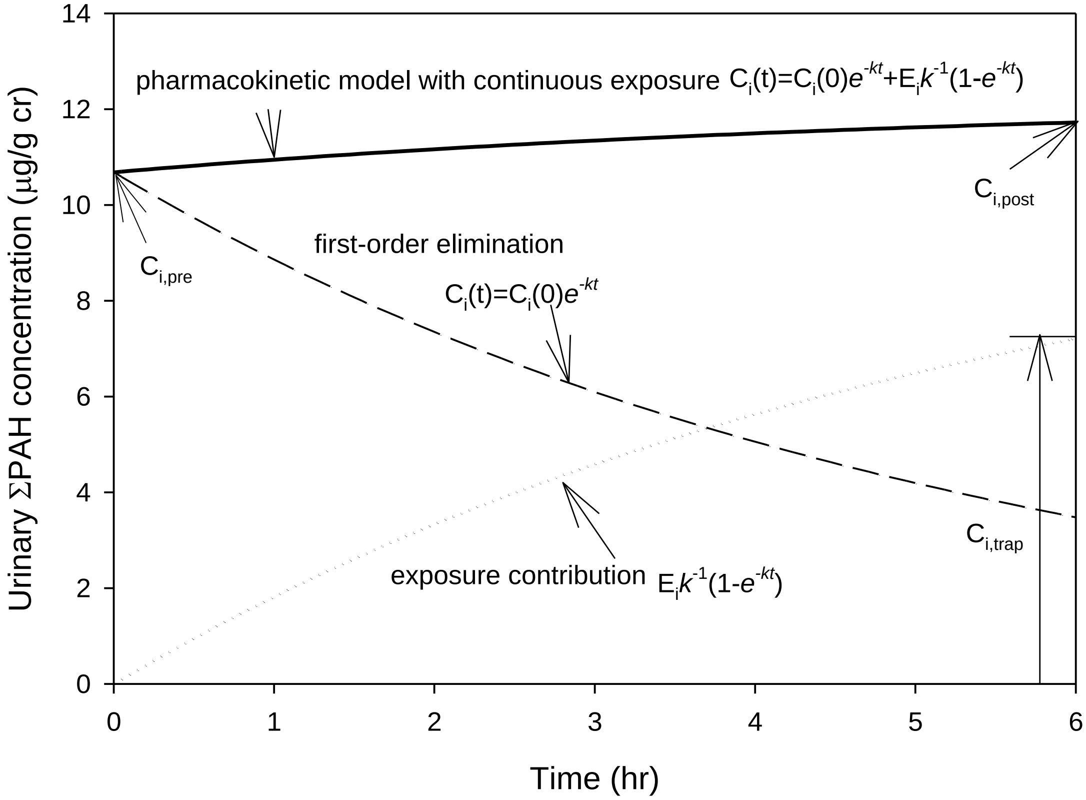


Figure 4.1. Taxi driver average urinary ∑OH-PAH concentration changes during the NM condition, plotted with the pharmacokinetic modeling method.

4.3.7 Data and statistical analysis

The urinary OH-PAH concentrations below detection limits (BDL) were calculated as half of the detection limit values. Calculated urinary OH-PAH increments due to the TRAP exposure during the monitored tests (OH-PAH_{trap}) were used as the surrogates of PAHs exposure during the monitored six hours. In order to normalize the variable distributions, all sampling results were log transformed. The Shapiro-Wilk test was conducted to ensure the transformed data were normally distributed before running the subsequent statistical tests and regression models.

Paired Student's t-tests were used to detect differences between interventions. Pearson's correlations and mixed effect linear regression models with random intercepts for each driver were used to test the associations between variables. The model can be expressed by the equation below:

$$y_{ij} = \alpha + \mu_i + \beta x_{ij} + \varepsilon_{ij} \tag{4.4}$$

Where i is the index of each driver, and j is the index of mitigation strategies. α and β are the fixed intercept and slope respectively, and μ_i is the random intercept of each driver under each mitigation condition. ε_{ij} is the residual. $PM_{2.5}$, UFP and $OH\text{-}PAH_{trap}$ were log transformed before fitting the model. The level of significance was taken as $p < 0.05$. SAS 9.4 software (SAS Institute, Cary, NC) was used for statistical analysis.

4.4 Results

4.4.1 On-road and in-cabin PM, and PAH exposures

The $PM_{2.5}$ and UFPs concentrations measured inside and outside the 17 taxis during the driving tests are summarized in Table 4.2. Because the start time, driving routes, and break times were well controlled, and the driving tests were conducted in three consecutive days for each driver, there is no significant difference detected of the on-road $PM_{2.5}$ and UFPs among different test conditions.

Under the “No Mitigation (NM)” condition, the vehicle ventilation and window position were controlled by the drivers. The sampling results reflect their exposures to UFP, $PM_{2.5}$, and PAHs under normal working conditions. The geometric mean $PM_{2.5}$ and UFPs levels inside the tested 17 taxis were 19 µg / m^3 and 1.40 x 10^4 particles / cm^3 respectively. The model calculated increments due to traffic exposure for ∑OH-NAP, ∑OH-FLU, ∑OH-PHE and 1-OH-PYR were 4.69, 0.51, 0.15, and 0.05 µg/g creatinine (cr), respectively (Table 4.2).

Compared with the NM condition, closing windows (WC) changed the in-cabin geometric mean $PM_{2.5}$ and UFP by -5% and 5% respectively, but the differences were not statistically significant. WC also did not significantly reduce the taxi driver urinary OH-PAH$_{trap}$ of any individual PAH species (Table 4.2). However, when HECA filters were used under the WC condition (WC+HECA), compared with NM, the geometric mean $PM_{2.5}$, UFP, and OH-PAH$_{trap}$ levels were reduced by 37 %, 47 %, and 5 %, respectively, and the reduction of $PM_{2.5}$ and UFP concentrations were statistically significant ($p < 0.05$) (Tables 4.2). This indicates that, as a mitigation strategy, WC+HECA is effective in reducing PM levels inside these tested 17 taxi vehicles.

Table 4.2 also shows that, the geometric mean post-test MDA concentrations were similar between NM and WC (1% difference), but was reduced by 17% under the WC+HECA condition. Although the reduction is not statistically significant, the trend among the three mitigation conditions was similar with the trends of in-cabin $PM_{2.5}$ and UFP concentrations, suggesting the

MDA changes might be linked with $PM_{2.5}$ and UFP reduction. This hypothesis is tested in the following section.

Table 4.2. Summary of taxi drivers' exposures under different test conditions (geometric mean, interquartile range, and percentage of change from no mitigation (NM))

Mitigation / Exposure	NM[a]	WC[b]	WC+HECA[c]
On-road $PM_{2.5}$ ($\mu g/m^3$)	31 (20, 53)	28 (20, 40) (-10%)[d]	29 (24, 33) (-6%)
In-cabin $PM_{2.5}$ ($\mu g/m^3$)	19 (15, 22)	20 (16, 20) (5%)	12 (10, 16) * (-37%)
On-road UFP ($x10^4$ cm^{-3})	2.71 (2.44, 3.02)	2.72 (2.29, 3.46) (0%)	2.70 (2.34, 3.55) (-0%)
In-cabin UFP ($x10^4$ cm^{-3})	1.40 (1.13, 1.97)	1.33 (1.07, 1.77) (-5%)	0.74 (0.59, 1.02) * (-47%)
$\sum$OH-NAP_{trap} (μg/g cr)	4.69 (2.30, 8.96)	3.47 (1.99, 5.38) (-26%)	4.40 (2.50, 6.47) (-6%)
$\sum$OH-FLU_{trap} (μg/g cr)	0.51 (0.29, 0.71)	0.40 (0.30, 0.74) (-22%)	0.48 (0.33, 0.60) (-6%)
$\sum$OH-PHE_{trap} (μg/g cr)	0.15 (0.08, 0.24)	0.12 (0.07, 0.20) (-20%)	0.18 (0.14, 0.20) (20%)
1-OH-PYR_{trap} (μg/g cr)	0.05 (0.02, 0.10)	0.05 (0.02, 0.10) (0%)	0.07 (0.05, 0.10) (40%)
Pre-test MDA (μg/g cr)	20.80 (20.53, 54.18)	10.74 (9.23, 40.95) (-48%)	27.14 (13.49, 56.39) (30%)
Post-test MDA (μg/g cr)	31.91 (22.78, 47.25)	31.73 (22.87, 43.90) (-1%)	23.68 (9.99, 55.37) (-17%)

a. No mitigation.
b. Window closed.
c. High efficiency cabin air filter.
d. Numbers in parenthesis indicate percentage of change from NM.

* indicate significance of paired t-test ($p < 0.05$) compared with NM.

4.4.2 Associations among PM, OH-PAHs and MDA

The in-cabin geometric mean $PM_{2.5}$ concentrations were not significantly correlated with UFP or any calculated $OH\text{-}PAH_{trap}$ levels. However, the geometric mean UFP concentrations were correlated significantly with $1\text{-}OH\text{-}PYR_{trap}$ levels ($p < 0.05$). This is consistent with the fact that pyrene has a vapor pressure of 4.5×10^{-6} mmHg at 25ºC,0 and possible to be found in particle phase in ambient air, and PAHs are more abundant in UFPs than in $PM_{2.5}$ [83]. For each OH-PAH species, significant correlations between pre- and post- test levels were detected ($p < 0.05$, Table 4.3), suggesting significant impacts of PAHs baseline levels on the post-test OH-PAHs. This justifies using the calculated $OH\text{-}PAH_{trap}$ as the surrogate of the 6-hr driving test PAH exposure instead of the post-test urinary OH-PAHs.

Unlike the urinary OH-PAHs, there is no significant correlation detected between the pre- and post- test MDA concentrations. However, the post-test MDA levels showed the same trend with PM and $OH\text{-}PAH_{trap,}$ that the lowest geometric mean was observed under WC+HECA (Table 4.2). These results also indicate that the pre-test MDA levels didn’t affect the post-test MDA levels significantly, but the TRAP exposures probably did. Thus, the association between PM and PAH exposures and post-test MDA concentrations was examined.

Table 4.3. Summary of OH-PAH changes under different test conditions (mean ± SD and reduction from NM) with the pharmacokinetic model. No significance detected from paired t-test comparing with no mitigation (NM). **unit**

Mitigation	NM	WC	WC+HECA
Increment			
1-OH-NAP	0.47±0.58	0.25±0.25 (-47%)	0.31±0.29 (-34%)
2-OH-NAP	1.55±1.26	1.23±1.09 (-21%)	1.11±0.80 (-28%)
∑OH-NAP	2.02±1.48	1.48±1.29 (-27%)	1.42±1.10 (-30%)
2-OH-FLU	0.70±0.69	0.45±0.21 (-36%)	0.50±0.25 (-29%)
3-OH-FLU	0.15±0.16	0.11±0.07 (-27%)	0.10±0.07 (-33%)
9-OH-FLU	0.96±0.70	0.75±0.58 (-22%)	0.79±0.41 (-18%)
∑OH-FLU	1.81±1.47	1.33±0.69 (-27%)	1.39±0.76 (-23%)
1-OH-PHE	0.23±0.28	0.15±0.07 (-35%)	0.21±0.12 (-9%)
2-OH-PHE	0.11±0.09	0.08±0.05 (-27%)	0.10±0.06 (-9%)
3-OH-PHE	0.17±0.20	0.12±0.07 (-29%)	0.13±0.08 (-24%)
∑OH-PHE	0.51±0.44	0.35±0.16 (-31%)	0.44±0.22 (-14%)
1-OH-PYR	0.28±0.43	0.18±0.13 (-36%)	0.21±0.21 (-25%)
Pre-test			
1-OH-NAP	3.48±5.22	2.30±2.71 (-34%)	2.24±3.04 (-36%)
2-OH-NAP	5.86±4.86	5.71±4.65 (-2%)	5.13±4.33 (-12%)
∑OH-NAP	9.34±8.85	8.02±4.99 (-14%)	7.37±6.20 (-21%)
2-OH-FLU	0.36±0.48	0.22±0.10 (-44%)	2.71±3.89 (-26%)
3-OH-FLU	0.16±0.15	0.11±0.07 (-31%)	0.18±0.33 (8%)
9-OH-FLU	0.47±0.34	0.42±0.22 (-11%)	0.48±0.41 (1%)
∑OH-FLU	1.00±0.87	0.73±0.29 (-26%)	0.92±1.08 (-8%)
1-OH-PHE	0.19±0.18	0.16±0.14 (-15%)	0.16±0.12 (-15%)
2-OH-PHE	0.08±0.08	0.05±0.03 (-29%)	0.07±0.09 (-13%)
3-OH-PHE	0.13±0.18	0.08±0.05	0.10±0.15

		(-37%)	(-20%)
∑OH-PHE	0.39±0.43	0.29±0.20 (-25%)	0.33±0.32 (-17%)
1-OH-PYR	0.14±0.12	0.13±0.09 (-5%)	0.12±0.10 (-13%)
Post-test			
1-OH-NAP	2.94±3.85	1.78±1.91 (-40%)	1.97±2.19 (-33%)
2-OH-NAP	5.80±4.53	5.05±4.63 (-13%)	4.46±3.03 (-23%)
∑OH-NAP	8.74±6.74	6.82±4.61 (-22%)	6.44±3.82 (-26%)
2-OH-FLU	0.31±0.32	0.20±0.08 (-36%)	0.23±0.16 (-26%)
3-OH-FLU	0.15±0.14	0.10±0.06 (-33%)	0.13±0.14 (-13%)
9-OH-FLU	0.43±0.28	0.37±0.24 (-14%)	0.39±0.22 (-9%)
∑OH-FLU	0.89±0.64	0.67±0.32 (-25%)	0.75±0.46 (-15%)
1-OH-PHE	0.17±0.17	0.13±0.08 (-24%)	0.15±0.07 (-12%)
2-OH-PHE	0.06±0.05	0.05±0.02 (-17%)	0.06±0.04 (0%)
3-OH-PHE	0.11±0.14	0.08±0.04 (-27%)	0.09±0.06 (-18%)
∑OH-PHE	0.35±0.36	0.26±0.12 (-24%)	0.30±0.16 (-14%)
1-OH-PYR	0.15±0.19	0.12±0.08 (-20%)	0.12±0.09 (-20%)

a. Window closed.

b. HECA filter in use.

c. Percentage of change from non-mitigation.

A mixed effect linear model was used to investigate the association between the taxi drivers' post-test urinary MDA and their PM/PAH exposures. Other factors that might affect the MDA levels, such as temperature, relative humidity, and weekday / weekend, were also included to the initial model, but the results showed that they were not significant factors, and the associations between exposures and the MDA were not affected. Thus the final models only include exposures and individual effects. Linear regression coefficients and their confidence intervals are summarized in Figure 4.2 to show the significance of the correlations at $\alpha = 0.05$ level. The taxi driver post-test urinary MDA levels were observed to have significant positive correlations with their in-cabin $PM_{2.5}$, and marginally significant positive correlations with their in-cabin UFP levels ($p = 0.07$) (Figure 4.2), which indicate that the oxidative stress might have occurred after the exposures.

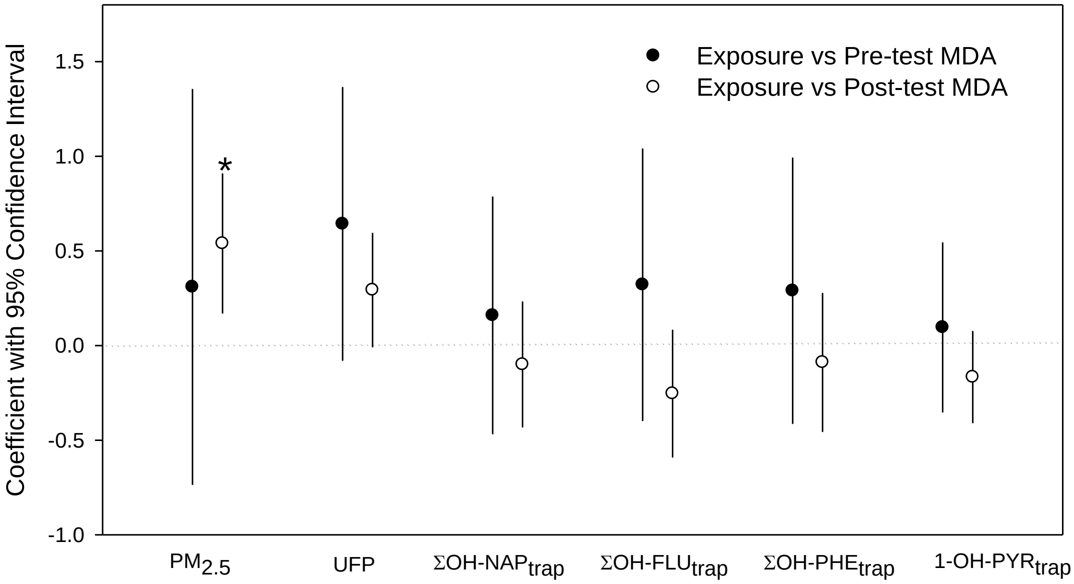


Figure 4.2. Correlations between pre- and post-test urinary MDA and PM/PAH exposures. * indicates significant correlations ($p < 0.05$), error bars indicate 95% confidence intervals.

4.5 Discussion

4.5.1 Taxi driver PM and PAH exposures

The $PM_{2.5}$ and UFP levels measured inside taxis under NM were on the similar levels with those measured inside private vehicles [20, 73]. However, since taxi drivers, on average, drive for 12 hours on each working day, which composes of 50% of their daily activities, their total exposures to roadway $PM_{2.5}$ and UFPs are about six times higher than other Southern California regular commuters, given that an average commuter in Southern California spends 7.2% of his/her day driving in traffic [84].

Urinary 1-OH-PYR is a widely used biomarker for human total PAH exposure assessments [85, 86]. The American Conference of Governmental Industrial Hygienist (ACGIH) has adopted the 1-OH-PYR as the Biological Exposure Determinant of the total PAHs in the Biological Exposure Indices (BEI) in 2009 [87]. In this study, urinary 1-OH-PYR concentration was used to compare the taxi drivers with other population. The results are illustrated in Figure 4.3. The median concentration of 1-OH-PYR of this taxi driver group was about 125% higher than the U.S. general population median from National Health and Nutrition Examination Survey (NHANES), 50% higher than a group of University of California Los Angeles (UCLA) students [41], but lower than taxi drivers or other occupational groups in Thailand and China [42, 88-90].

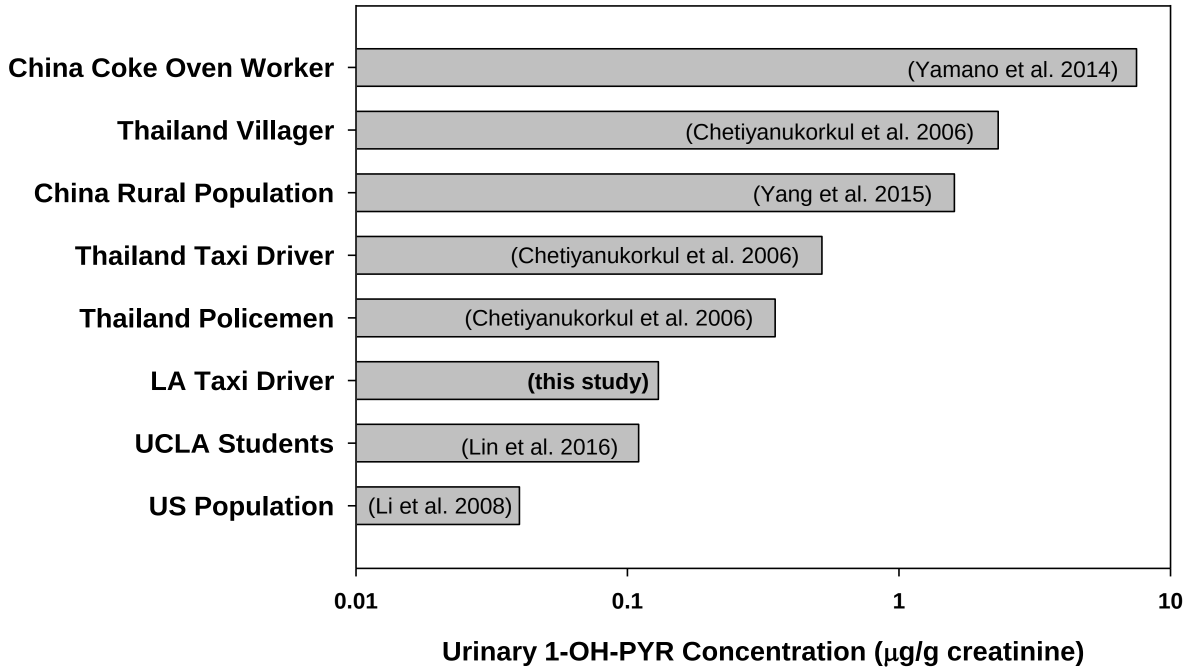


Figure 4.3. Comparison of Los Angeles taxi driver urinary 1-PYR concentrations with literature data from U.S. population (NHANES), UCLA students, and some other occupational groups.

4.5.2 Exposure mitigation

In this study, only 2-ring (NAP), 3-ring (FLU and PHE), and 4-ring (PYR) PAHs were measured by their urinary monohydroxylated metabolites. In all collected urine samples, on average, the two OH-NAPs (2.28 and 5.20 µg/g cr) were most abundant, followed by the three OH-FLUs (0.13-0.42 µg/g cr), the three OH-PHEs (0.06-0.15 µg/g cr), and the 1-OH-PYR (0.13 µg/g cr). The sum of the two OH-NAPs (∑OH-NAP), the only di-cyclic aromatic hydrocarbon metabolites, on average, contributed 83% to the ∑OH-PAH in mass. This is comparable with the 81.8% in the U.S. population from the NHANES results [90]. NAP, FLU, and PHE are mostly in gaseous phase in ambient air, while the PYR is mainly gaseous under relatively high ambient temperature [91]. The results show that 1-OH-PYR only composed of about 1% of the total analyzed OH-PAHs in mass. The recorded sampling temperature during the field sampling time ranged from 13.7 ºC to 34.9 ºC with an average of 27.8 ºC. This relatively high ambient temperature made the ambient PYR more likely to be found in the gaseous phase [92].

Compared with in-cabin $PM_{2.5}$ and UFP, the reduction of PAH exposure by WC+HECA was not significant, which is probably due to the gaseous nature of the measured PAHs. The HECA filter was less effective for gaseous PAHs than particles. Notably, the other PAH species in the particle phase (more than 4 rings) are also likely to be at high levels on roads and cause health risks [93]. Therefore, the results of this study only indicate that the effect of HECA filters was limited for gas phase PAHs, but might be still effective for higher molecular weight PAH reduction, which was not measured in this study.

4.5.3 PM / PAH exposures and health effects

The significant positive correlations between the in-cabin PM exposures and the post-test urinary MDA observed in this study support the previous findings that that PM exposures induce systematic oxidative stress in human. The possible pathways linking the PM exposures and urinary MDA generation include: (i) activation of leukocyte NADPH oxidase and myeloperoxidase [94]; (ii) interference with normal mitochondrial functions [95]; and (iii) depletion of antioxidant capacities [96]. Some chemical compositions of $PM_{2.5}$ and UFPs can also contribute to the oxidative stress through other pathways. In addition, the decrease of post-test MDA level under WC+HECA suggests the potential health benefit of the tested mitigation strategy.

Previous studies have reported significant association between urinary OH-PAHs and MDA [42, 70]. Such an association was not observed in this study. This is probably due to the differences in the study design. In this study, the urinary MDA levels were measured immediately after the exposures, reflecting the acute health effects, whereas most other studies on PAHs and oxidative stress association were based on long term observations. For example, one longitudinal study found significant association between urinary OH-PAHs and MDA based on samples collected over several months [41]. Different from the PM, the PAHs' capacity of inducing ROS depends on their relatively longer and more complex metabolizing processes. Given that the median half-lives of the analyzed PAHs in human body range from 2.5 to 6.1 hours, the insignificant association between OH-PAH$_{trap}$ and MDA in our study is possibly due to the fact that the post-test samples collected right after the 6-hr driving cannot reflect the MDA generation induced by the 6-hr PAH exposures. Nevertheless the difference in our results between PAHs and PM suggest different mechanisms in inducing oxidative stress.

There are several limitations of this study. First, 17 taxis and drivers were a small sample size. However, the study power was increased with repeated-measures study design, enabling each subject to serve as his own control. Second, no environmental PAH data were collected during the tests because of the instrument restrictions. Instead, urinary metabolites and a pharmacokinetic model were used to calculate the exposure surrogates, which may induce uncertainties. Specifically, only population mean elimination rates were available and used to calculate the OH-PAH$_{trap}$, and individual variations in PAH metabolism among the drivers were not taken into consideration. However, this interference from individual variations, could be at least partially controlled with the repeated measures design. The elimination rates in the literature from both dietary and inhalation intake are compared in Table 4.4. Although the calculated OH-PAH$_{trap}$ values were different between dietary and inhalation routes, the association between the OH-PAH$_{trap}$ and MDA remains insignificant. Finally, the concentration of MDA could also be influenced by factors other than the PM through inhalation, such as other inhalable pollutants and diet. However, the post-test MDA seems mainly affected by TRAPs because pre- and post- test MDA concentrations were not correlated.

To our knowledge, this is the first intervention study using repeated-measures design to examine the health effects of TRAPs on taxi drivers. The positive association between the in-cabin PM levels and the drivers' urinary MDA concentrations shows that oxidative stress is a possible contributor of the adverse health effects from the TRAP exposures.

Table 4.4. Comparison of calculated OH-PAH$_{\text{trap}}$ with dietary and inhalation intake half-lives

		Literature 1 **(Ref. of this study)** Li et. al. [82]	**Literature 2** Motorykin et. al. [97]	**Literature 3** **(Smoker study)** St Helen et. al. [98]
Intake route		***dietary***	***dietary***	***inhalation***
Reported half-life (hr) / elimination rate (k)	1-NAP	4.3 / 0.16	3.4 / 0.21	_8.6 / 0.08*_
	2-NAP	2.5 / 0.27	2.4 / 0.28	9.4 / 0.07
	2-FLU	2.9 / 0.24	2.6 / 0.26	4.1 / 0.17
	3-FLU	6.1 / 0.11	7.0 / 0.10	8.2 / 0.08
	9-FLU	3.1 / 0.23	1.7 / 0.41	_6.2 / 0.11*_
	1-PHE	5.1 / 0.14	3.1 / 0.22	_10.2 / 0.07*_
	2-PHE	3.9 / 0.18	3.7 / 0.19	_7.8 / 0.09*_
	3-PHE	4.1 / 0.17	2.6 / 0.27	_8.2 / 0.08*_
	1-PYR	3.9 / 0.18	4.4 / 0.16	6.0 / 0.12
Calculated OH-PAHtrap				
$\sum$OH-NAP$_{\text{trap}}$ (μg/g cr)	NM	4.69	5.27	2.24
	WC	3.47	4.06	0.63
	WC+HECA	4.40	3.94	1.39
$\sum$OH-FLU$_{\text{trap}}$ (μg/g cr)	NM	0.51	0.57	0.45
	WC	0.40	0.46	0.35
	WC+HECA	0.48	0.50	0.43
$\sum$OH-PHE$_{\text{trap}}$ (μg/g cr)	NM	0.15	0.20	0.10
	WC	0.12	0.17	0.08
	WC+HECA	0.18	0.19	0.10
1-OH-PYR$_{\text{trap}}$ (μg/g cr)	NM	0.05	0.05	0.02
	WC	0.05	0.04	0.01
	WC+HECA	0.07	0.03	0.03

*estimated values from Literature 1 by doubling the half-lives.

5 CONCLUSIONS

Previous epidemiological and toxicological studies have presented strong evidence supporting adverse health effects of TRAPs. Previous studies have investigated passenger vehicle and school bus drivers' and commuters' exposures to TRAPs and the possible mitigation strategies. Taxi drivers and passengers, who are exposed to higher levels of TRAPs, have been sparingly studied. PM concentrations in regular passenger vehicles can be 10 times higher than the ambient levels and taxis are usually leakier due to greater wear and tear. Taxi drivers work 72 hours a week driving on roadways on average, and they potentially have high total exposures to TRAPs. This dissertation study aimed to investigate the UFP and $PM_{2.5}$ levels on-road and inside taxis, along with the taxi vehicle I/O ratios, under four different ventilation and mitigation conditions. The taxi drivers' urinary OH-PAH and MDA concentrations and the association with their PM exposures were also studied.

The on-road UFP and $PM_{2.5}$ modeling results show that, it is practical to use meteorological, traffic and spatial parameters to predict arterial road UFP and $PM_{2.5}$ concentrations. GAMs and sGAMs explained higher percentages of variance among the three model types (i.e., MLR model, GAM and sGAM), whereas the MLR models generated least over fitting results. The performance of the GAMs was not improved by the sGAMs. Higher percentages of variance of the arterial road measurements can be explained by the developed models than the percentages of variance of freeway and local surface street measurements with the same type of model. Spatial features such as land use and altitude were not predictors in the best performed arterial road GAMs.

On arithmetic average, the Los Angeles taxi drivers are exposed to arithmetic means of 1.46 x 10^4 particles / cm^3 and 26 µg / m^3 $PM_{2.5}$ while driving on the roadways without mitigation. Although he results show that, the $PM_{2.5}$ and UFP levels measured inside taxis under NM were similar to those measured inside private vehicles, considering their long driving time, their total exposures to roadway $PM_{2.5}$ and UFPs are about six times higher than other Southern California regular commuters. The four ventilation and mitigation conditions of NM, WC, WC+HECA and HECA reduced the on-road UFP levels by 40%, 35%, 53%, and 49%, and reduced on-road $PM_{2.5}$ levels by 25%, 34%, 48%, and 39%, respectively. UFP and $PM_{2.5}$ levels inside taxis as well as their I/O ratios were significantly different under each of the four conditions. UFP and $PM_{2.5}$ had the lowest I/O ratios, 0.47 and 0.52, under the WC+HECA condition. Simply closing taxi windows (WC) but using OA ventilation mode did not effectively reduce UFP and $PM_{2.5}$ I/O ratios in Toyota Prius taxis. However, using a HECA filter made the Toyota Prius taxis have the lowest UFP and $PM_{2.5}$ I/O ratios across all tested taxi models.

Compared with in-cabin $PM_{2.5}$ and UFP, the reduction of PAH exposure by WC+HECA was not significant, which is probably due to the gaseous nature of the measured PAHs. The HECA filter was less effective for gaseous PAHs than particles. The taxi driver post-test urinary MDA levels were observed to have significant positive correlations with their in-cabin $PM_{2.5}$, and marginally significant positive correlations with their in-cabin UFP levels ($p = 0.07$), which indicate that the oxidative stress might have occurred after the exposures, and the WC+HECA strategy was helpful with the oxidative stress reduction.

The results suggest that controlling window position and ventilation setting, and improving cabin air filters are promising methods to reduce $PM_{2.5}$ and UFP levels inside taxis.

6. APPENDICES

Taxi Driver Screening Survey Form

1. Name : ________________________________ ________________________________
(First Name) (Last Name)

2. Would you be interested in participating in an air pollution study that includes compensation for your time? ☐ YES ☐ NO

3. Contact information (phone number and/or e-mail):_______________________________________

4. What is your age? ☐ < 25 ☐ 25-35 ☐ 36-45 ☐ 46-55 ☐ >55

5. Gender: ☐ Male ☐ Female

6. a) Height: _______ ft _______ inches b) Weight: __________ lbs

7. What is the highest level of education you have completed?
   ☐ Less than high school ☐ Some high school ☐ High School diploma or GED
   ☐ Some college or associate's degree ☐ Bachelor's degree (College)
   ☐ Some graduate school ☐ Graduate degree (M.A., Ph.D. etc.)

8. What is your race/ethnicity?
   ☐ Black ☐ Hispanic/Latino ☐ White/Non-Hispanic
   ☐ Asian ☐ Native American ☐ Pacific Islander
   Other, *please specify:* _________________________

9. Marital Status: ☐ Married/Living with partner ☐ Divorced/Widowed ☐ Never married

10. Number of children who are financially dependent on you: _________________________________

11. What are the make, model, and year of your car?

Make: _______________ Model: _____________ Year: _________ Plate number: _________________

12. Are you the owner of your taxi or do you lease from a taxi company?
    ☐ Owner ☐ Lease ☐ Other: ______________________________

13. How long have you been a taxi driver?
    ☐ < 6 Months ☐ 6 – 12 Months or __________ Years

14. Typically, when do you start and end your work day? Start: ________ $\frac{\text{AM}}{\text{PM}}$ End: _________ $\frac{\text{AM}}{\text{PM}}$

15. How many days do you typically work as a taxi driver each week? ___________DAYS

16. How much time do you spend driving on freeways during a work day?
    ☐ < 3 hours ☐ 3 – 6 hours ☐ 6 – 9 hours ☐ > 9 hours

17. How often do you drive with your windows down?

☐ < 25% of time ☐ 25 – 50% ☐ 50 – 75% ☐ > 75%

18. Do you smoke?

☐ Never ☐ Quit ☐ Smoke 1 cigarette per day or less
☐ Smoke 2 – 10 cigarettes per day ☐smoke ½ - 1 pack cigarettes per day
☐Smoke more than 1 pack cigarettes per day

19. Have you ever been diagnosed with any of the following conditions?

a) Heart condition? ☐ No ☐ Yes, please specify: _______
b) High blood pressure (hypertension) ☐ No ☐ Yes, please specify: _______
c) A respiratory condition (i.e. asthma, COPD, etc.)? ☐ No ☐ Yes, please specify: _______

20. Have you missed work or been hospitalized in the past year because of your

a) Heart condition? ☐ Yes, for how long: ________________________________ ☐ No
b) High blood pressure ☐ Yes, for how long: ________________________________ ☐ No
c) Respiratory condition ☐ Yes, for how long: ________________________________ ☐ No
d) Any other condition ☐ Yes, for how long: ________________________________ ☐ No

21. Do you currently take medication for any of the following:

a) Heart condition ☐ No ☐ Yes
b) Hypertension? ☐ No ☐ Yes
c) High cholesterol? ☐ No ☐ Yes
d) Diabetes? ☐ No ☐ Yes

22. In general, would you say your health is:

☐ Excellent ☐ Ve**ry good** ☐ Good ☐ Fair ☐ Poor

23. On average during the past year, how many days a week have your work-outs met both criteria:
* 30 minutes or more in duration each time
* Medium (work up a sweat and slight heart rate increase) to vigorous intensity (work up a good sweat and rapid heart rate increase)

☐ 0 days/week ☐ 1 day/week ☐ 2 days/week ☐ 3 or more days/week

24. How often do you find your work stressful?

☐ Always ☐ Often ☐ Sometimes ☐ Hardly ever ☐ Never

25. Do you have health insurance? ☐ No ☐ Yes

26. Do you plan to quit working as a taxi driver within the next 6 months? ☐ No ☐ Yes

Thank you for your time. Return the form at the <u>holding lot exit</u> and get a ***free gift !***

Taxi driver questionnaire

Taxi Driver Air Pollution Exposure Questionnaire

Section 1

All personal information will be kept confidential.

Prefix: Food and drink information

During the last three days:

1. Did you cook at home in the last three days?

   No.

   Yes. How long have you spent cooking? (in hours)

2. What is the main type of fuel used for cooking? (circle one)

   A. Gas; B. Electric; C. Others (specify)

3. Did you barbecue in the last three days?

   No.

   Yes. How long have you spent to barbecue? (in hours)

4. Did you have barbecue in the last three days?

   No.

   Yes. How much barbecue have you had? (in pounds)

5. Did you have baked meat/food in the last three days?

   No.

   Yes. How much baked meat/food have you had? (in pounds)

6. What was your main food in the last three days? (circle one or more)

   A. Hamburger; B. Piza; C. Rice;
   D. Noodle; E. Others (specify)

7. Was anyone smoking indoors around you?

No.

Yes. How long have you stayed with them each day? (in hours)

Part A. Demographic Information

A1. Name: ____

(Last/Family Name) (First/Given Name) (Middle Initial)

A2. Phone numbers:

Cell: ______________________________________

Home: ______________________________________

Other: ______________________________________

A3. Age: ____________

A4. If married or living with a partner, does your partner/spouse work outside the home?

☐ Full-time ☐ Part-time ☐ Does not work outside the home

A5. Number of children living with you at home: Under 5 years ________ Over 5 years________

A6. What was your income in the last 12 months?

Personal Income	Household Income (all income earners)
☐ 1) Less than $15,000	☐ 1) Less than $15,000
☐ 2) $15,000-$35,000	☐ 2) $15,000-$35,000
☐ 3) $36,000-$55,000	☐ 3) $36,000-$55,000
☐ 4) $56,000-$75,000	☐ 4) $56,000-$75,000
☐ 5) Over $75,000	☐ 5) Over $75,000

A7. During the past 7 days, even if it was not typical for you, how much total time did you spend on exercise?

☐ None ☐ 0-0.5 hour ☐ 0.5– 1 hour ☐ 1 -3 hours ☐>3 hours

A8. During the past 7 days, even if it was not typical for you, did you consume any alcoholic drinks?

☐ No

☐ Yes. Then please specify the type of drink and the total amount consumed in the past 7 days:

A8a. Beer:

☐ None ☐ less than 1 can ☐1-3 cans ☐ more than 3 cans

A8b. Wine:

☐ None ☐ less than 1 wine glass ☐1-3 wine glass ☐ more than 3 cans

A8c. Hard Liquor (such as Gin, Rum, Vodka, Whiskey):

☐ None ☐less than 1 shot glass ☐1-3 shot glass ☐ more than 3 shot glass

A9a. How many hours of sleep do you typically get in every 24 hours? _______hours
A9b. How big do you consider your problems are, in the past 7 days, with:

		None **(1)**	**Small** **(2)**	**Moderate** **(3)**	**Very Big** **(4)**
a)	Falling asleep at beginning	☐	☐	☐	☐
b)	Waking up during the sleep	☐	☐	☐	☐
c)	Waking up too early	☐	☐	☐	☐
d)	Not feeling rested by your sleep	☐	☐	☐	☐

	Strongly Disagree (1)	**Disagree (2)**	**Agree (3)**	**Strongly Agree (4)**
A10a. I eat more of my favorite foods to make me feel better, under stressful events.	☐	☐	☐	☐
A10b. I eat more than I usually do, under stressful events.	☐	☐	☐	☐

A11a. How many servings of food do you eat (per day) that are high in fiber, such as whole grain bread, high fiber cereal, fresh fruits or vegetables? (a serving size: 1 slice bread, ½ cup vegetables, 1 medium fruit, ¾ cup cereal)

☐ (1) 5-6 servings/day

☐ (2) 3-4 servings/day

☐ (3) 1-2 servings/day

☐ (4) never/rarely

Taxi Driver Air Pollution Exposure Questionnaire

Section 2

All personal information will be kept confidential.

Prefix: Food and drink information

During the past 24 hours:

1. Did you cook at home?

   No.

   Yes. How long have you spent in cooking? (in hours)

2. What is the main type of fuel used for cooking? (circle one)

   A. Gas; B. Electric; C. Others (specify)

3. Did you barbecue ?

   No.

   Yes. How long have you spent to barbecue? (in hours)

4. Did you have barbecue?

   No.

   Yes. How much barbecue have you had? (in pounds)

5. Did you have baked meat/food?

   No.

   Yes. How much baked meat/food have you had? (in pounds)

6. What was your main food in the last three days? (circle one or more)

   A. Hamburger; B. Piza; C. Rice;
   D. Noodle; E. Others (specify)

7. Was there anyone else smoke indoor around you?

No.

Yes. How long have you stayed with them each day? (in hours)

Part B. Clinical Information: Respiratory

The following questions are about respiratory or chest symptoms. If you are in doubt whether the answer is Yes or No, answer No.

Respiratory Symptoms

B1. In the past 12 months, have you had a cough on most days or nights of the week during at least three months in a row? ("Most" means at least 4 days or nights per week)

☐ No ☐ Yes

B2. Have you had a cough on getting up or first thing in the morning on most mornings (at least 4 mornings per week) for at least three months in a row?

☐ No ☐ Yes

B3. In the past 12 months, have you brought up phlegm from your chest on most days or nights of the week during at least three months in a row? ("Most" means at least 4 days or nights per week)

☐ No ☐ Yes

B4. Have you brought up phlegm on getting up or first thing in the morning on most mornings (at least 4 per week) for at least three months in a row?

☐ No ☐ Yes

B5. Have you ever had an attack of wheezing or whistling in your chest that made you feel short of breath?

☐No ☐Yes

B6. In the last 12 months, does your chest ever sound wheezy or whistling when you have a cold?

☐No ☐Yes

B7. In the last 12 months, have you been awakened from sleep either by coughing (apart from a cough associated with a cold or chest infection) or by shortness of breath or a feeling of tightness in your chest?

☐No ☐Yes

B8. When you are near animals (such as cats, dogs, or horses) or near feathers (including pillows, quilts or comforters) or in a dusty or moldy part of the house, do you ever:

B8a. start to cough, wheeze, feel short of breath, or feel a tightness in your chest?

☐No ☐Yes

B8b. get a runny or stuffy nose or start to sneeze, or get itching or watering eyes?

☐No ☐Yes

B9. When you are near trees, grass, or flowers, or when there is a lot of pollen in the air, do you ever:

B9a. start to cough, wheeze, feel short of breath, or feel a tightness in your chest?

☐No ☐Yes

B9b. get a runny or stuffy nose, start to sneeze, or get itching or watering eyes?

☐No ☐Yes

B10. Have you ever had allergen skin testing?

☐No ☐Yes

B11. Do you have chronic sinusitis?

☐No ☐Yes

B12. When you exercise or exert yourself or when the air is cold, do you ever start to cough, wheeze, feel short of breath, or feel tightness in your chest?

☐No ☐Yes

B13. Are you troubled by shortness of breath when hurrying on level ground or walking up a slight hill?

☐No ☐Yes

B14. During the past 12 months, about how many days of work did you miss because of respiratory illnesses or symptoms?

☐None ☐ 1-5 ☐6-15 ☐16 or more

B15. During the past 12 months, have you had respiratory symptoms (cough, phlegm, wheeze, or shortness of breath) that got better on weekends, vacations, or other times when you were away from your current job? If more than one current job, consider the job you spend the most time doing.

☐No ☐Yes ☐Don't know ☐Not applicable

Respiratory Conditions

B16. Have you ever had asthma?

☐No ☐Yes ☐Don't know

B17. Was it diagnosed by a doctor or other health professional?

☐No ☐Yes ☐Don't know

B18. In the past 12 months, have you received medical treatment, taken medications or used an inhaler for asthma?

☐No ☐Yes

B19. In the past 12 months, have you received medical treatment, taken medications or used a nasal spray for hay fever?

☐No ☐Yes

B20. Has a doctor ever told you that you had pneumonia or bronchopneumonia?

☐No ☐Yes ☐ Don't know

B21. Has a doctor ever told you that you had chronic bronchitis?

☐No ☐Yes ☐ Don't know

B22. Has a doctor ever told you that you had COPD (chronic obstructive pulmonary disease) or emphysema?

☐No ☐Yes ☐ Don't know

B23. In the past 12 months, have you received medical treatment, taken medications or used an inhaler for COPD or emphysema?

☐No ☐Yes

Respiratory Family History

B24. Has a doctor ever said that these relatives had an attack of asthma?

B24a. Mother

☐ No or Don’t know ☐ Yes

B24b. Father

☐ No or Don’t know ☐ Yes

B24c. Sibling(s) (Don’t consider half-brothers and half-sisters.)

☐ No or Don’t know ☐ Yes

B25. Has a doctor ever said that these relatives had chronic bronchitis, COPD, or emphysema?

B25a. Mother

☐ No or Don’t know ☐ Yes

B25b. Father

☐ No or Don’t know ☐ Yes

B25c. Sibling(s) (Don’t consider half-brothers and half-sisters.)

☐ No or Don’t know ☐ Yes

B26. Has a doctor ever said that these relatives had hay fever (allergy involving the nose and/or eyes)?

B26a. Mother

☐ No or Don’t know ☐ Yes

B26b. Father

☐ No or Don’t know ☐ Yes

B26c. Sibling(s) (Don’t consider half-brothers and half-sisters.)

☐ No or Don’t know ☐ Yes

Part C. Clinical Information: Cardiovascular

Cardiovascular Symptoms

The following questions are about cardiovascular or chest symptoms. If you are in doubt whether the answer is Yes or No, answer No.

C1. What is your Total Cholesterol level: ☐ Don’t know ☐ Yes, I know. It is ___ mg/dL

C2. What is your HDL Cholesterol level: ☐ Don’t know ☐ Yes, I know. It is ___ mg/dL

C3. Do you have Atrial Fibrillation?

☐No ☐Yes ☐ Don’t know

C4. Do you take medications for high blood pressure?

☐No ☐Yes

C5. Do you have a history of smoking in your life time?

☐No ☐Yes

C6. Do you have a history of diabetes?

☐ No ☐ Yes ☐ Don’t know

C7. Have you experienced in the past year, chest pain described as:

C9a. during exercise

☐ No ☐ Yes

C9b. Brief duration (2-15min)

☐ No ☐ Yes

C9c. Relieved promptly by rest or TNG/Nitroglycerin/Nitro

☐ No ☐ Yes

C9d. Retrosternal (behind your breast bone)

☐ No ☐ Yes

C9e. Radiating to jaw, neck or left arm

☐ No ☐ Yes

C9f. Absence of other cause

☐ No ☐ Yes

C8. Have you consulted a physician about your cardiovascular symptoms?

☐ No ☐ Yes

Cardiovascular Conditions

C9. Has any doctor or health professional told you that you have cardiovascular disease (i.e., high blood pressure, coronary artery disease, cardiac dysrhythmias)?

☐ No ☐ Yes

C10. In the past 12 months, have you received medical treatment, taken medications for heart disease?

☐ No ☐ Yes

Cardiovascular Family History

C12. Do any of your blood relatives have a history of heart disease?

C13a. Mother:

☐ No ☐ Yes ☐ Don't know

C13b. Father:

☐ No ☐ Yes ☐ Don't know

C13c. Sibling(s) (Don't consider half-brothers and half-sisters.)

☐ No ☐ Yes ☐ Don't know

Taxi Driver Air Pollution Exposure Questionnaire

Section 3

All personal information will be kept confidential.

Prefix: Food and drink information

During the past 24 hours:

8. Did you cook at home?

   No.

   Yes. How long have you spent in cooking? (in hours)

9. What is the main type of fuel used for cooking? (circle one)

   A. Gas; B. Electric; C. Others (specify)

10. Did you barbecue ?

    No.

    Yes. How long have you spent to barbecue? (in hours)

11. Did you have barbecue?

    No.

    Yes. How much barbecue have you had? (in pounds)

12. Did you have baked meat/food?

    No.

    Yes. How much baked meat/food have you had? (in pounds)

13. What was your main food in the last three days? (circle one or more)

    A. Hamburger; B. Piza; C. Rice;
    D. Noodle; E. Others (specify)

14. Was there anyone else smoke indoor around you?

No.

Yes. How long have you stayed with them each day? (in hours)

Part D. Exposure and Risk Factor Information

Occupational History

D1. What were your occupations before you started driving a taxi?

(1) ____ No. Years:________________

(2) ____ No. Years:________________

(3) ____ No. Years:________________

(4) ____ No. Years:________________

(5) ____ No. Years:________________

(6) ____ No. Years________________

D2. Did you have your respiratory symptoms/conditions before becoming a taxi driver?

☐ No ☐ Yes

D3. Did you begin to develop your cardiovascular symptoms before becoming a taxi driver?

☐ No ☐ Yes

D4. Did you begin to develop your respiratory symptoms/conditions after becoming a taxi driver?

☐ No ☐ Yes

D5. Did you begin to develop your cardiovascular symptoms after becoming a taxi driver?

☐ No ☐ Yes

D6. Did your respiratory symptoms/conditions worsen after becoming a taxi driver?

☐ No ☐ Yes

D7. Did you cardiovascular symptoms worsen after becoming a taxi driver?

☐ No ☐ Yes

D8. Have you ever experienced your respiratory symptoms during your work shift?

☐ No ☐ Yes

D9. Have you ever experienced your respiratory symptoms right after your work shift?

☐ No ☐ Yes

D10. Have you experienced your cardiovascular symptoms during your work shift?

☐ No ☐ Yes

D11. Have you experienced your cardiovascular symptoms right after your work shift?

☐ No ☐ Yes

D12. Do you experience high levels of stress due to your job as a taxi driver?

☐ No ☐ Yes

D13. Do you worry about your exposure to pollutants in the air when you are driving?

☐ No ☐ Yes

Current Home Environment

D14. During the last 12 months, has there been any flooding or water damage in your home?

☐ No ☐ Yes

D15. During the last 12 months, have you noted any mold or mildew on any surface, other than food, inside your home?

☐ No ☐ Don’t know ☐ Yes

D16. Is your house within 300 ft of a major freeway such as an interstate or state highway?

☐ No ☐ Yes

Part E. Work Characteristics

E1. How many miles did you drive last YEAR for work? ___________________________

E2. How many miles did you drive last WEEK for work?___________________________

E3. On a typical work shift, how many rest breaks do you take?___________________

E4. How many passenger-fares do you usually pick up in a typical work day?_______________

The following questions are about work stress. Please indicate the extent to which you agree or disagree (Check one box per question):

	Strongly Disagree (1)	**Disagree (2)**	**Agree (3)**	**Strongly Agree (4)**
E5. My job requires that I learn new things. (JCQ-SD)	☐	☐	☐	☐
E6. My job requires me to be creative. (JCQ – SD)	☐	☐	☐	☐
E7. I have an opportunity to develop my own special abilities. (JCQ – SD)	☐	☐	☐	☐
E8. On my job, I am given a lot of freedom to decide how I do my work. (JCQ – DA)	☐	☐	☐	☐
E9. I have a lot to say about what happens on my job. (JCQ – DA)	☐	☐	☐	☐
E10. My job requires working very fast. (JCQ – D)	☐	☐	☐	☐
E11. My job requires working very hard. (JCQ – D)	☐	☐	☐	☐
E12. My job requires lots of mental effort. (JCQ)	☐	☐	☐	☐
E13. My job requires lots of physical effort. (JCQ)	☐	☐	☐	☐
E14. My job often involves lifting loads weighing 50 pounds or greater.	☐	☐	☐	☐
E15. My job_often involves crouching, stooping, or kneeling.	☐	☐	☐	☐
E16. My job often requires sitting for long periods of time.	☐	☐	☐	☐
E17. I am not asked to do an excessive amount of work.(JCQ –D)	☐	☐	☐	☐
E18. I have enough time to get the job done. (JCQ –D)	☐	☐	☐	☐
E19. I am free from conflicting demands others make. (JCQ –D)	☐	☐	☐	☐

E20. My job security is good. (ERI – R)	☐	☐	☐	☐
E21. I have constant time pressure due to a heavy work load (ERI – E)*	☐	☐	☐	☐
E22. Over the past years, my job has become more and more demanding.(ERI – E)	☐	☐	☐	☐
E23. I am treated unfairly at work. (ERI – R)	☐	☐	☐	☐
E24. I have many interruptions and disturbances while performing my job (ERI – E)	☐	☐	☐	☐
E25. I receive the respect and prestige I deserve at work. (ERI – R)	☐	☐	☐	☐
E26. My salary/income is adequate.(ERI – R)	☐	☐	☐	☐

E27. I have "co-workers" who I work with regularly ☐ Yes ☐ No (If NO, go to Q. 24)

If yes, please answer the following:	**Strongly Disagree (1)**	**Disagree (2)**	**Agree (3)**	**Strongly Agree (4)**
a) My coworkers are friendly	☐	☐	☐	☐
b) My coworkers are helpful in getting the job done	☐	☐	☐	☐
c) I am exposed to hostility or conflict from my coworkers	☐	☐	☐	☐

Work-Family Balance	**Strongly Disagree (1)**	**Disagree (2)**	**Agree (3)**	**Strongly Agree (4)**
1. The demands of my work interfere with my home and family life.	☐	☐	☐	☐
2. The amount of time my job takes up makes it difficult to fulfill family responsibilities.	☐	☐	☐	☐
3. Due to work-related duties, I have to make changes to my plans for family activities.	☐	☐	☐	☐
4. The demands of my family or spouse/partner interfere with work-related activities.	☐	☐	☐	☐
5. I have to put off doing things at work because of demands on my time at home.	☐	☐	☐	☐
6. My home life interferes with my responsibilities at work such as getting to work on time, accomplishing daily tasks, and working.	☐	☐	☐	☐

Please answer the following questions about YOUR INTERACTIONS WITH THE GENERAL PUBLIC WHILE DRIVING. How frequently do you find yourself doing the following while working with the public (Check one box per question):

EMOTIONAL LABOR	**Never/ Not at all (1)**	**Rarely/ Once in a while (2)**	**Some-times (3)**	**Often/ Most of the Time (4)**	**Always/ Constantly (5)**
7. I have to deal with people who are difficult or disrespectful.	☐	☐	☐	☐	☐
8. My work is emotionally demanding.	☐	☐	☐	☐	☐
9. I make an effort to actually feel the emotions I need to display toward the public.	☐	☐	☐	☐	☐
10. I hide my genuine emotions about things that happen when working.	☐	☐	☐	☐	☐
11. I put on the appearance of being calm and professional for my job, even when not feeling that way.	☐	☐	☐	☐	☐
12. I easily contain my emotions and express myself calmly and professionally to the public	☐	☐	☐	☐	☐

General Health Status

BURNOUT – Emotional Exhaustion: Please read the following items and decide if you ever feel this way about your job. If you have never had this feeling, check box for “Never.” If you have had this feeling, indicate how often you feel this way:

	Never (1)	**A few times a year (2)**	**Once a month (3)**	**A few times a month (4)**	**Once a week (5)**	**A few times a week (6)**	**Every day (7)**
13. I feel emotionally drained from my work.	☐	☐	☐	☐	☐	☐	☐
14. I feel used up at the end of the workday.	☐	☐	☐	☐	☐	☐	☐
15. I feel tired when I get up in the morning and have to face another day on the job.	☐	☐	☐	☐	☐	☐	☐
16. Working all day is really a strain for me.	☐	☐	☐	☐	☐	☐	☐
17. I feel burned out from my work.	☐	☐	☐	☐	☐	☐	☐

General Health Questionnaire (GHQ12): We want to know how your health has been in general <u>over the last 4 weeks</u>. Please read the questions below and each of the four possible answers. Check the response that best applies to you.

Have you recently:

	Not at all (0)	**No more than usual (1)**	**Rather more than usual (2)**	**Much more than usual (3)**
18. Lost much sleep over worry?	☐	☐	☐	☐
19. Felt constantly under strain?	☐	☐	☐	☐
20. Felt you couldn't overcome your difficulties?	☐	☐	☐	☐
21. Been feeling unhappy or depressed?	☐	☐	☐	☐
22. Been losing confidence in yourself?	☐	☐	☐	☐
23. Been thinking of yourself as a worthless person?	☐	☐	☐	☐

Have you recently:	**More than usual (0)**	**Same as usual (1)**	**Less than usual (2)**	**Much less than usual (3)**
24. Been able to concentrate on what you're doing?	☐	☐	☐	☐
25. Felt that you are playing a useful part in things?	☐	☐	☐	☐
26. Felt capable of making decisions about things?	☐	☐	☐	☐
27. Been able to enjoy your normal day to day activities?	☐	☐	☐	☐
28. Been able to face up to your problems?	☐	☐	☐	☐
29. Been feeling reasonably happy, all things considered?	☐	☐	☐	☐

Taxi Driver Air Pollution Exposure Questionnaire

Section 4

All personal information will be kept confidential.

Prefix: Food and drink information

During the past 24 hours:

15. Did you cook at home?

    No.

    Yes. How long have you spent in cooking? (in hours)

16. What is the main type of fuel used for cooking? (circle one)

    A. Gas; B. Electric; C. Others (specify)

17. Did you barbecue ?

    No.

    Yes. How long have you spent to barbecue? (in hours)

18. Did you have barbecue?

    No.

    Yes. How much barbecue have you had? (in pounds)

19. Did you have baked meat/food?

    No.

    Yes. How much baked meat/food have you had? (in pounds)

20. What was your main food in the last three days? (circle one or more)

    A. Hamburger; B. Piza; C. Rice;
    D. Noodle; E. Others (specify)

21. Was there anyone else smoke indoor around you?

No.

Yes. How long have you stayed with them each day? (in hours)

The following is a voice-recording interview:

Script:

Today is (month, date). I am (Segovia or Nu). Now I am interviewing taxi driver Mr. (name).

Mr. (name), please take a moment to answer following questions about your work:

1. What is the best part(s) about your job?

____

____

____

2. What is the most difficult aspect of your job?

____

____

____

3. What are some actions that could be taken to improve working life at your current job?

____

____

____

4. Additional comments about your work and would you be willing to participate in future studies?

____

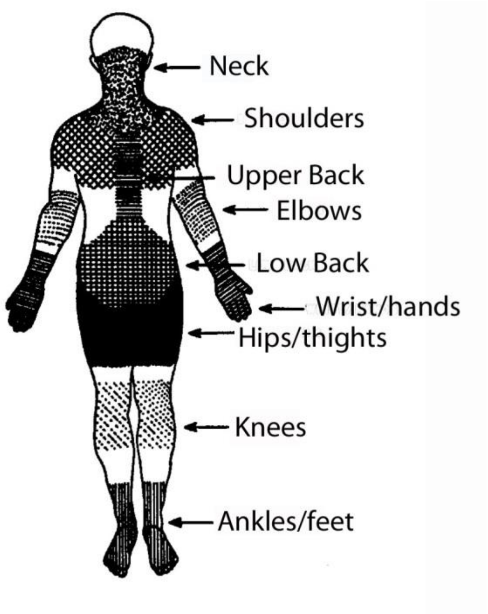
____

Part F. Standardized Nordic Questionnaire

ow to answer:

Please answer by putting a cross in the appropriate box-one cross for each question. You may be in doubt as to how to answer, but please do your best anyway. Please answer every question, even if you have never had trouble in any part of your body.

In this picture you can see the approximate position of the parts of the body referred to in the questionnaire. Limits are not sharply defined, and certain parts overlap. You should decide for yourself in which part you have or have had your troubles (if any).



		To be answered only by those who have had trouble			
Have you at any time during the last 12 months had trouble (ache, pain, discomfort) in:		Have you at any time during the last 12 months been prevented from doing your normal work (at home or away from home) because of the trouble?		Have you had trouble at any time during the last 7 days?	
Neck					
1 ☐ No	2 ☐ Yes	1 ☐ No	2 ☐ Yes	1 ☐ No	2 ☐ Yes
Shoulders					
1 ☐ No	2 ☐ Yes in the right shoulder				
	3 ☐ Yes in the left shoulder				
	4 ☐ Yes in both shoulder	1 ☐ No	2 ☐ Yes	1 ☐ No	2 ☐ Yes
Elbows					
1 ☐ No	2 ☐ Yes in the right elbow				
	3 ☐ Yes in the left elbow				
	4 ☐ Yes in both elbow	1 ☐ No	2 ☐ Yes	1 ☐ No	2 ☐ Yes
Wrists/hands					
1 ☐ No	2 ☐ Yes in the right wrist/hand				
	3 ☐ Yes in the left wrist/hand				
	4 ☐ Yes in both writs/hand	1 ☐ No	2 ☐ Yes	1 ☐ No	2 ☐ Yes
Upper back					
1 ☐ No	2 ☐ Yes	1 ☐ No	2 ☐ Yes	1 ☐ No	2 ☐ Yes
Low back (small of the back					
1 ☐ No	2 ☐ Yes	1 ☐ No	2 ☐ Yes	1 ☐ No	2 ☐ Yes
One or both hips/thighs					
1 ☐ No	2 ☐ Yes	1 ☐ No	2 ☐ Yes	1 ☐ No	2 ☐ Yes
One or both knees					
1 ☐ No	2 ☐ Yes	1 ☐ No	2 ☐ Yes	1 ☐ No	2 ☐ Yes
One or both ankles/feet					
1 ☐ No	2 ☐ Yes	1 ☐ No	2 ☐ Yes	1 ☐ No	2 ☐ Yes

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Characterization of Ultrafine Particles and Other Traffic Related Pollutants near Roadways in Beijing

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ABSTRACT

Developing countries, such as China, are facing serious air pollution issues due to fast economic development. In this study, traffic related air pollutants, including number concentration of ultrafine particles (UFPs, diameter < 100 nm), mass concentrations of $PM_{2.5}$ and black carbon (BC) were measured near the Peking University (PKU) campus in Beijing in December 2011. Data were collected concurrently at a roadway site and on PKU campus. Meteorological data were collected at approximately 40 meters northeast from the roadway sampling site. The traffic density was determined from recorded video footage. Roadside UFP and $PM_{2.5}$ concentrations were not significantly higher than on campus. A statistically significant Pearson's correlation of 0.75 was found between BC and $PM_{2.5}$ mass concentrations. No apparent correlation was found between wind speed and UFP number concentrations, but strong log-decay correlations were found between wind speed and $PM_{2.5}$ ($R^2 = 0.80$). There were three days during the measurements when both $PM_{2.5}$ mass concentrations and UFP number concentrations were higher at the campus site than at the roadway site. This suggests there were potential local emission sources on campus. Temporal profile of UFPs at the campus site peaked around lunch and dinner time, suggesting emissions from the surrounding restaurants and cafeteria that used Chinese-style cooking might have contributed to the observed $PM_{2.5}$ and UFP levels on campus.

Keyword: Ultrafine particles; $PM_{2.5}$; Black carbon; Beijing; Roadway; Cooking emissions.

INTRODUCTION

Air pollution studies conducted in the US and Europe have shown a consistent relationship between increases in particulate matter (PM) exposure and increases in human mortality and morbidity. In developing countries, such as China, the rapid industrialization and urbanization have led to huge increases of PM emissions. Beijing is one of the megacities in China with a population of 20.69 million. According to 2012 Beijing Environmental Statement, the 2012 annual average PM_{10} (particles with aerodynamic diameter ≤ 10 µm) mass concentration was 109 $µg/m^3$, which exceeded the China Grade II National Ambient Air Quality Standards (70 $µg/m^3$) by 56% (Beijing Municipal Environmental Protection Bureau, 2013).

PM pollution has been linked to adverse health outcomes, particularly respiratory and cardiovascular diseases (Schulz *et al.*, 2005; Leitte *et al.*, 2009; Brook *et al.*, 2010). These diseases are prevalent in China, where air pollution levels are often orders of magnitude higher than those in developed countries (Zhang *et al.*, 2011). Because PM deposition and related health effects depend on the particle size, $PM_{2.5}$ (fine particles with aerodynamic diameter ≤ 2.5 µm) and ultrafine particles (UFP; diameter < 100 nm) have been associated more strongly with health risk than PM_{10} (Politis *et al.*, 2008). Recent studies have shown that UFP may be particularly damaging to human health due to their ability to penetrate deeper into the lung and induce oxidative stress in deep lung tissue (Oberdorster, 2001; Donaldson *et al.*, 2002; Politis *et al.*, 2008).

Previous studies conducted in the United States and some European countries have demonstrated a link between vehicle traffic density and UFP, $PM_{2.5}$, and Black Carbon (BC) levels (Harrison *et al.*, 1997). However, a study in Beijing suggested that vehicular emissions were not the dominant factor affecting the total UFP variations in Beijing urban area (Shi *et al.*, 2007). In that study, the air sampling was conducted on Tsinghua University campus and no comparable data were collected near roadway to directly reflect traffic emissions. In this study, UFP and $PM_{2.5}$ were measured concurrently at a near-road site and a campus site to evaluate the impact of traffic on UFPs in Beijing. Other emission sources that might also affect UFP variations were identified and discussed.

METHODS

Sampling Sites

Data were collected at two stationary sampling sites located on and near the Peking University (PKU) campus in Beijing, China (Fig. 1). Fig. 1(c) illustrates the orientations of the major roads and the locations of the sampling sites. The campus sampling site was located on the roof of a six-story building on PKU main campus (Site A on Fig. 1(c)), which is about 20 meters above the ground. It was located at the east central part of PKU campus surrounded by research and office buildings, student residence halls, and dining facilities, which are all about two to six stories in height. The roadside sampling site was located at the entrance to the PKU School of Chemistry, which is outside the PKU East Gate to the main campus. This sampling site was set up at the gate guard station right outside the entrance facing Chengfu Road (Site B on Fig. 1(c)). The sampling inlets were about 1 meter above the ground. Site B was approximately 25 meters from the center of an intersection of North Zhongguancun Road and ChengfuRoad. The highest building within 300 meter distance to the sampling sites is the nine-story Founder Building located at the southeast corner of the intersection. Other buildings along the two main roads in this area are all about five to seven stories in height and are located about 20 meters from the paved pedestrian's walkways of the roads. The North Zhongguancun Road and Chengfu Road are busy arterial roads with two to three vehicle lanes in each direction in addition to bicycle lanes and paved pedestrians walkways. Traffic on these roads includes light duty gasoline vehicles as well as diesel trucks and buses. In general, the North Zhongguancun Road runs from north to south with subway line operating underground and Chengfu Road runs from PKU east gate to east. Fig. 1 also shows the potential cooking emission sources such as restaurants and cafeterias on PKU campus. The majority of these emission sources were located approximately 200–500 meters to the east of sampling site A.

Meteorological data were collected by the PKU Automatic Meteorology Station, which is located on the third floor (~10 meter above ground) at the north building of PKU School of Physics. It is approximately 40 meters northeast from sampling site B. Data generated from this weather station were used to represent the local meteorological pattern over the sampling period.

Instrumentation and Data Collection

At Site A, a TSI water based condensation particle counter (CPC; Model 3786, TSI Inc., Shoreview, MN) was used to measure total UFP concentrations roughly from 5 nm to a few micrometers. Because in urban environments, UFPs usually constitute > 90% of total particle number concentrations, we used UFP to refer to CPC readings in this study. A TSI DustTrak photometer (Model 8520, TSI Inc., Shoreview, MN) with a 2.5 µm inlet impactor was used to measure $PM_{2.5}$ mass concentrations. The water based CPC was placed in an enclosed room on the roof due to its temperature sensitive nature. To prevent particle loss in the sampling line to the electrostatic charge, TSI conductive tubing was used and

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extended through a window to the outside ambient air. The DustTrak was placed on the roof

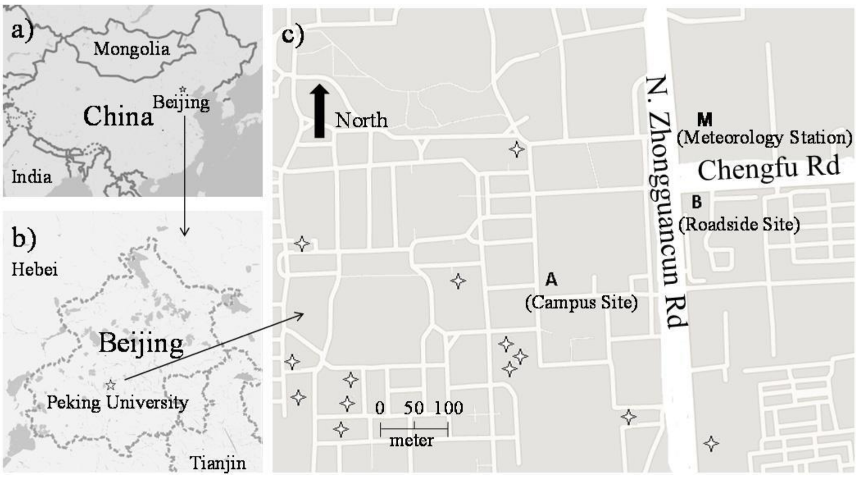

Fig. 1. Maps of stationary sampling sites on PKU campus (Site A) and near roadways (Site B). Cooking emission sources were illustrated by four-point stars on c).

directly. The reading of DustTrak was generally higher than gravimetric measurements compared with the United States Environmental Protection Agency (USEPA) designated Federal Reference Method (Chung *et al.*, 2001; Yanosky *et al.*, 2002). Therefore, the DustTrak data were calibrated against simultaneous gravimetric measurements of $PM_{2.5}$ on PKU campus. A factor of 2.4 was achieved and used for DustTrak data correction, which was consistent with data reported in previous studies (Yanosky *et al.*, 2002; Zhang and Zhu, 2010).

At Site B, a second set of water based CPC (Model 3785, TSI Inc., Shoreview, MN) and DustTrak (Model 8520, TSI Inc., Shoreview, MN) with a 2.5 µm inlet impactor were used to measure UFPs and $PM_{2.5}$, respectively. In addition, a Magee Scientific Aethalometer (Model AE-42, Magee Scientific Corporation, Berkeley, CA) was used to measure BC mass concentrations. The water based CPC was placed in a gate guard station and TSI conductive tubing was used to connect to the sampling inlet and extended through a window to outside ambient air. The DustTrak and Aethalometer were placed outdoor.

These two sets of instruments were operating simultaneously from 8 am to 9 pm during the sampling campaign. The CPCs were set to take readings every second. The DustTraks and the Aethalometer were set to measure at 1-min intervals. All instruments were within the manufacturer annual calibration. All the instruments were co-located before and after the field campaign to make sure there were no significant differences between the two sets of instruments in terms of real time readings. Data were downloaded each day immediately after the sampling was finished.

For traffic density, video recordings were made using a commercial camera with a tripod at the guard station. The camera was pointing towards the T intersection of North Zhongguancun Road and Chengfu Road. Recordings were made for 5 minutes every 15 minutes throughout the sampling period. Thus, for one hour of sampling, a total of 20 minutes recording were made. The number of vehicles was then counted per minute from the recording.

RESULTS AND DISCUSSION

UFP, $PM_{2.5}$, and BC Concentrations

The basic descriptive statistics for UFPs, $PM_{2.5}$, and BC concentrations are shown in Table 1. All of these measured concentrations show high variability. Comparing the data collected from Sites A and B, the average UFP number concentrations and the average $PM_{2.5}$ mass concentrations did not show significant differences. However, the standard deviation of UFP concentrations was much lower at Site A than at Site B (1.20×10^4 vs. 3.23×10^4 #/cm^3). This indicates more stable UFP emission sources at Site A while the Site B UFP concentrations are primarily influenced by intermittent traffic emissions. The standard deviations of $PM_{2.5}$ mass concentrations were similar at both sampling sites. $PM_{2.5}$ was a little but not significantly higher (91.70 vs. 80.31 µg/m^3) at Site A than at Site B. The measured average BC mass concentration at Site B was 4.70 µg/m^3 with a standard deviation of 5.41 µg/m^3 during the total 14 days of the sampling campaign. The high variability of BC mass concentrations at Site B is also likely due to the large number of diesel buses used for public transportation in Beijing.

At Site B, the average percentage of BC in $PM_{2.5}$ was 5.9%. Literature values of BC and BC/$PM_{2.5}$ ratios are listed in Table 3 and presented in Fig. 2. The reported average BC and $PM_{2.5}$ concentrations at different sampling sites and at different sampling time are linearly correlated with an R^2 value of 0.87 (Fig. 2). The BC/$PM_{2.5}$ ratio observed in this study was close to or slightly lower than most of those reported in previous studies conducted in Beijing (Dan *et al.*, 2004; Yang *et al.*, 2005; Zhao *et al.*, 2013), as well as at other global sites. A one-year study at an urban site in Helsinky, Finland in 2000 through 2001 showed the BC in $PM_{2.5}$ was about 14 ± 8% (Viidanoja *et al.*, 2002). Another study conducted in New York state also showed a BC/$PM_{2.5}$ mass concentration ratio of about 15% (Schwab *et al.*, 2004). A previous study in Beijing in 2001 showed BC in $PM_{2.5}$ was about 6.8% (He *et al.*, 2001). This is likely because our data were mostly collected during daytime (from 8 am to 9 pm). A previous study conducted in Beijing urban and rural areas in 2009 indicated that heavy-duty diesel vehicle (HDDV) flow usually peaked around 240 vehicles/hour at midnight, but ranged from 0– 20 vehicles/hour between 6 am and 9 pm. Accordingly, the diurnal BC concentrations reached the highest value of 29 µg/m^3 at midnight in winter (Song *et al.*, 2013). This is because due to the local traffic control rules in Beijing, HDDVs were only allowed to operate during night time (11 pm to 6 am next day) in the urban area. Thus, in this study, we were not able to capture the highest BC traffic emissions from those HDDVs. Since BC mass concentration is an indicator of combustion sources including diesel traffic, the lower percentage of BC in $PM_{2.5}$ mass concentration in Beijing indicates that the diesel traffic may not be a predominant factor contributing to the total $PM_{2.5}$ concentrations in winter in Beijing during the daytime.

Table 1. Basic descriptive statistics of UFP, $PM_{2.5}$, and BC.

	UFP (#/cm^3)		$PM_{2.5}$ (µg/m^3)		BC (µg/m^3)
	Site A	Site B	Site A	Site B	Site B
Mean	3.62×10^4	3.61×10^4	93.10	80.30	4.70
Std Dev	1.20×10^4	3.23×10^4	91.71	87.02	5.41
Geometric Mean	3.43×10^4	3.23×10^4	55.54	49.03	3.01
Geometric Std Dev	1.38	1.59	2.87	2.66	2.62
Min.	1.00×10^3	5.40×10^3	3.00	9.00	0.19

Max.	1.13×10^5	2.20×10^5	5.85×10^2	5.43×10^2	29.43

Correlations between UFP, PM$_{2.5}$, and BC

The UFP number concentrations and PM$_{2.5}$ and BC mass concentrations were averaged at 30 minute intervals for all correlation analysis. Table 2 summarized Pearson's correlation coefficients among different pollutant measurements at both Site A and Site B. At both sampling sites, Pearson's correlations between UFPs and PM$_{2.5}$ mass concentrations were not statistically significant. The UFP concentrations at Sites A and B were not significantly correlated either, despite that the average concentrations were within the similar range. This indicates the UFP sources at the two sampling sites were different albeit the two sites were only ~200 m apart. In contrast, Site B BC correlated with PM$_{2.5}$ mass concentrations at Sites A and B, so did PM$_{2.5}$ mass concentrations at both sampling sites (Table 2).

As shown in Fig. 2, data from previous studies have a strong spatial correlation between PM$_{2.5}$ and BC mass concentrations. In the present study, a similarly high temporal correlation was observed between roadside BC and PM$_{2.5}$ at both sampling sites. This result is not surprising since previous studies conducted in Beijing showed PM$_{2.5}$ and BC share some common sources from coal and fuel burning, especially in the winter months (Dan *et al.* , 2004). Atmospheric scales of air pollutant transport can be roughly categorized into microscale (0–100 meters), mesoscale (tens to hundreds of kilometers), synoptic scale (hundreds to thousands of kilometers) and global scale (> 10^3 kilometers). Both PM$_{2.5}$ and BC have been classified as mesoscale air pollutants that can travel tens to hundreds of kilometers (Krudysz *et al.*, 2008; Wang *et al.* , 2011). This is because their atmospheric lifetime is fairly long so that they can be transported and dispersed within a large region (Cape *et al.*, 2012; Chen *et al.*, 2013). When the atmospheric conditions are stagnant, PM$_{2.5}$ and BC concentrations may also increase if local emission sources such as traffic exist. When the atmospheric PM$_{2.5}$ levels are high, it generates the well-known "Beijing Haze" phenomenon, which strongly affects visibility in the urban areas. Once PM$_{2.5}$ and BC are emitted, their concentrations are affected by meteorological conditions such as wind speeds and directions. Furthermore, the vast majority of BC aerosol is in the fine particle size range (i.e., less than 2.5 µm) and typically comprises 5–10% of total PM$_{2.5}$ (Table 3).

Meteorological Effects on UFP and PM$_{2.5}$

In Beijing, local meteorology is often highly variable, which affects the local ambient air quality dramatically as reported in many previous studies (Feng *et al.*, 2005). Half-

Table 2. Pearson correlation coefficients among measured pollutants at campus site (A) and roadside site (B)

	UFP (A)	PM$_{2.5}$ (A)	UFP (B)	PM$_{2.5}$ (B)	BC (B)
UFP (A)	1.000(-)				
PM$_{2.5}$ (A)	0.176 (0.002)	1.000(-)			
UFP (B)	0.318 (< 0.001)	0.166 (0.046)	1.000(-)		
PM$_{2.5}$ (B)	0.012 (0.849)	0.824 (< 0.001)	0.037 (0.694)	1.000(-)	
BC (B)	0.150 (0.009)	0.871 (< 0.001)	0.008 (0.926)	0.753 (<0.001)	1.000(-)

Notes: P values are given in parentheses.

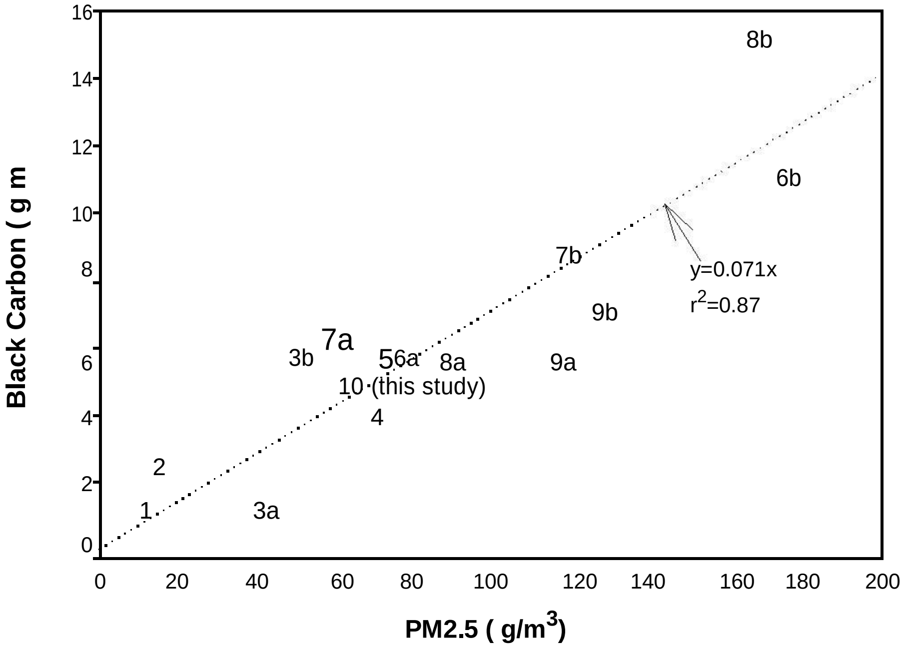

Fig. 2. Correlations between BC and PM$_{2.5}$ at different global sites in the literature. The study sites and related references

Table 3. Summary of literature values of BC and BC/$PM_{2.5}$ ratio.

Study ID	Location	Year	BC Average ($\mu g/m^3$) (S.D.)	BC/$PM_{2.5}$ (%)	Reference
1	Helsinki, Finland	2001–2002	1.2	14.0	Viidanoja *et al.* (2002)
2	Los Angeles, US	2011	2.5	20.6	Quiros *et al.* (2013)
3a	HongKong	2000–2001	Background: 1.36	3.2	Ho et al. (2002)
3b			Urban: 5.80	11.2	
4	Kaohsiung, Taiwan	1998–1999	4.0	5.9	Lin *et al.* (2001)
5	Pearl River Delta	2002	6.1	8.8	Cao *et al.* (2003)
6a	Beijing Urban	1999–2000	Summer: 6.3	8.3	He *et al.* (2001)
6b			Winter:11.1	6.3	
7a	Shanghai Urban	1999–2000	6.10	10.0	Yang *et al.* (2005)
7b	Beijing Urban	1999–2000	8.79	7.5	
8a	Beijing Urban	2001–2003	Summer: 5.7 (2.9)	6.3	Dan *et al.* (2004)
8b			Winter: 15.2 (11.1)	9.0	
9a	Beijing Urban	2009	Summer: 5.9 (1.2)	5.1	Zhao *et al.* (2013)
9b		2010	Winter: 7.1 (2.2)	5.6	
10	Beijing Urban	2011	Winter: 4.7 (5.4)	5.9	This study

hour wind speeds and directions during the 14-day sampling campaign were summarized in Fig. 3 where the size of the dots indicates time of day and the distance from the center indicates different wind speeds. During the sampling periods, approximately 70% of the time, the wind blew from northwest to southeast (270–360 degrees), under which the Site A was upwind and Site B was downwind of the roads.

Collected data showed wind directions didn't significantly affect $PM_{2.5}$ concentrations at both sites, BC concentrations at Site B, and UFP concentrations at Site B. Average UFP concentrations at Site A were slightly higher when the wind is from NW than other directions. Correlations between wind speeds and UFP and $PM_{2.5}$ are shown in Fig. 4. To simplify the wind direction effects on the associations between wind speed and pollutants, only upwind UFP and $PM_{2.5}$ data were used for Site A and downwind UFP and $PM_{2.5}$ data were used for Site B. Wind speed and UFP number concentration did not show any correlations (Figs. 4(a) and 4(b)), however, wind speed and $PM_{2.5}$ shows apparent log-decay correlations with R^2 values of 0.81 for the Site A and 0.80 for Site B (Figs. 4(c) and 4(d)). Higher wind speeds are associated with a fast drop in average $PM_{2.5}$ mass concentrations at both sampling sites. This is consistent with previous findings that $PM_{2.5}$ and BC share common regional sources such as biomass burning in winter in Beijing (Dan *et al.*, 2004; Yang *et al.*, 2005; Zhao *et al.*, 2013). They also had comparable atmospheric lifetime on the order of weeks which make their concentrations affected similarly by air movements such as convection (atmospheric stability) and advection (wind), especially when regional source dominates.

Compared with $PM_{2.5}$, UFP concentrations tend to be indicative of local emissions. UFP concentrations were found to drop sharply as the distance from a roadway is increased due to both particle coagulation and atmospheric dispersion (Zhu *et al.*, 2002). However, the effect of wind speed on UFPs in this study is different from studies conducted in the US, where increasing wind speeds were shown to be statistically associated with a decrease in UFP because of the dispersion effect from the air movement. In this study, the UFP concentrations remain relatively stable at different wind speeds. This is likely because in Beijing, extremely high $PM_{2.5}$ levels can reduce UFP lifetime (Shi *et al.*, 2007). In this study, much higher $PM_{2.5}$ concentrations were observed at lower wind speeds (Figs. 4(c) and 4(d)). These fine particles provided larger surface areas for UFP to deposit. In addition, these fine particles provide a condensation sink for semi-volatile vapors and hence particle formation through nucleation processes both in vehicle exhaust plumes and in the wider atmosphere is suppressed by high pre-existing particle loadings. When the wind speed increased, assuming the emission rates were similar for all UFP sources, a stronger atmospheric dispersion was expected to reduce UFP concentrations. On the other hand, at higher wind speed, $PM_{2.5}$ concentrations became much lower and the deposition surface for UFP became limited. These two competing processes may keep ambient UFP levels relatively stable with respect to wind speed in Beijing.

There is also a strong association ($R^2 = 0.96$) between wind speed and BC mass concentrations, with a profile similar to that between wind speed and $PM_{2.5}$. BC and $PM_{2.5}$ also share similar diurnal profile as shown in Fig. 5. This result is consistent with previous studies that have shown BC generally is enriched in the $PM_{2.5}$ size range (Chaloulakou *et al.*, 2003; Richmond-Bryant *et al.*, 2009).

Cooking Sources and Traffic Influence

Diurnal UFP, $PM_{2.5}$, and BC levels were analyzed against meteorological data. Wind speed was dichotomized into low wind (wind speed < 4.5 m/s) and strong wind (wind speed ≥ 4.5 m/s) as shown in Fig. 5. This results in about half and half data for each category. Under strong wind condition and when Site A was upwind of the roadways (wind is from 0 and 180 degrees), the UFP diurnal pattern shows bimodal at around 11 am and 6 pm (Fig. 5(b)). This result is consistent with previous study conducted on Tsinghua University campus, about 2 km from the sampling

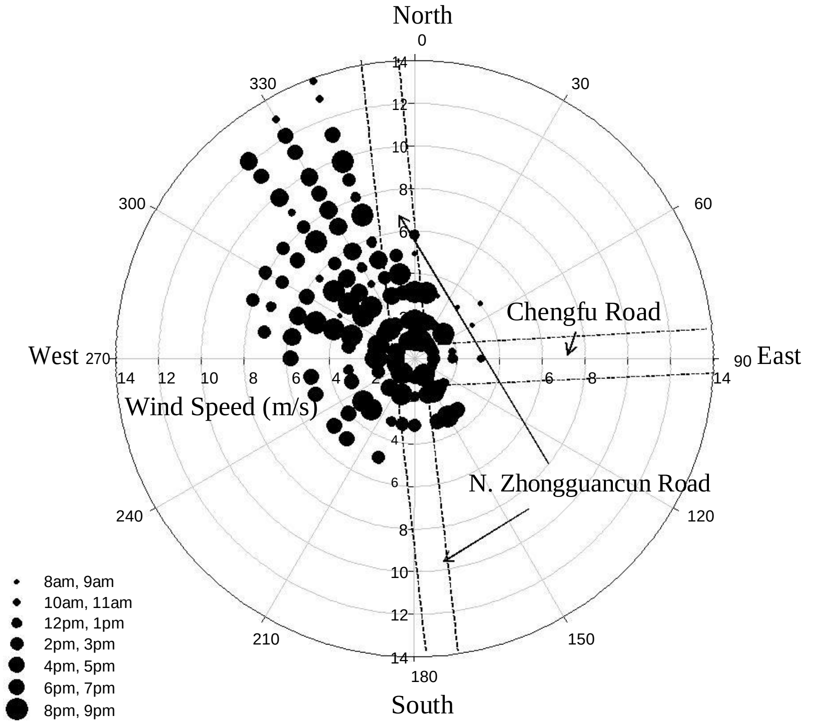

Fig. 3. Half-hour averaged wind speed and direction. Radius indicates wind speed and angle indicates wind direction, while 0 degree indicates north and 90 degree indicates east. Size of the dots on the graph indicates time of day with the smallest one representing 8–9 am.

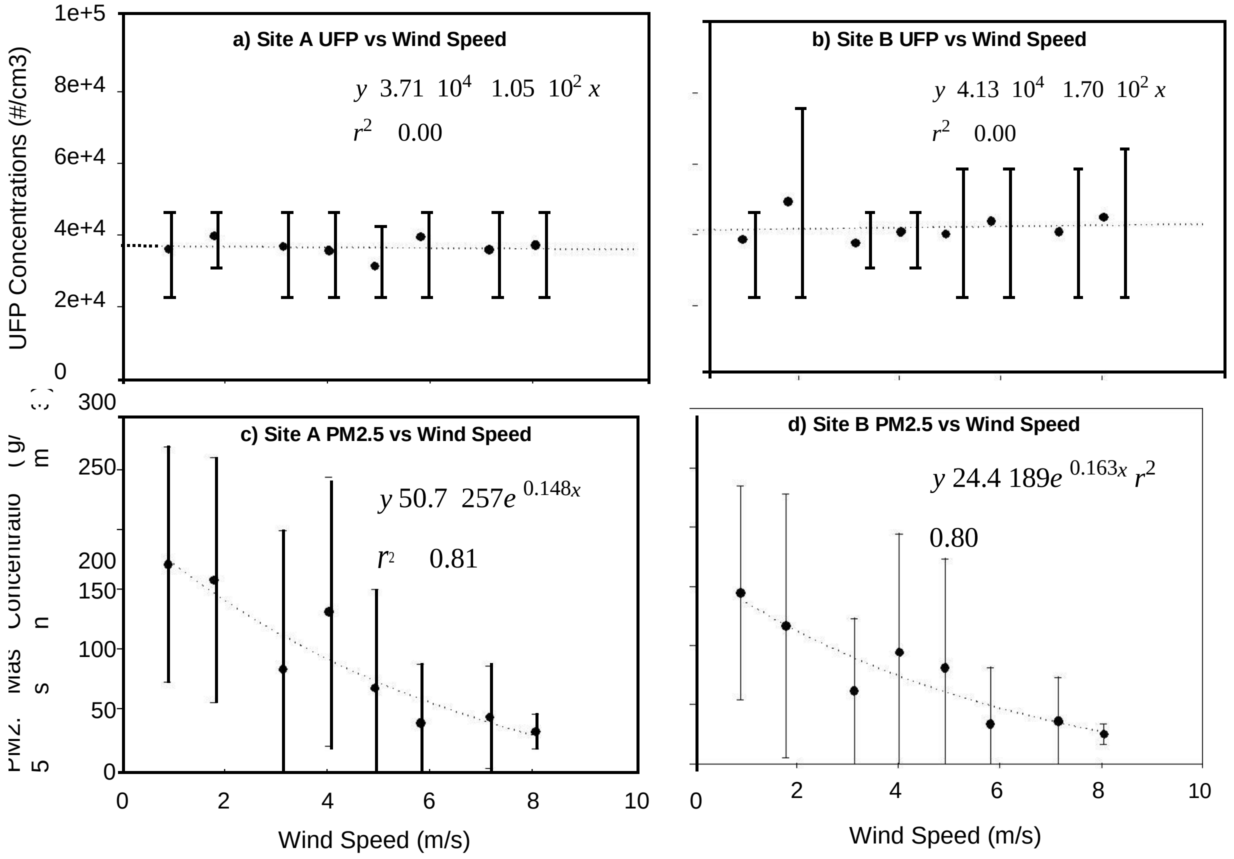

Fig. 4. The effects of wind speed on UFP number concentrations and $PM_{2.5}$ mass concentrations at Sites A and B. Error bars stand for one standard deviation.

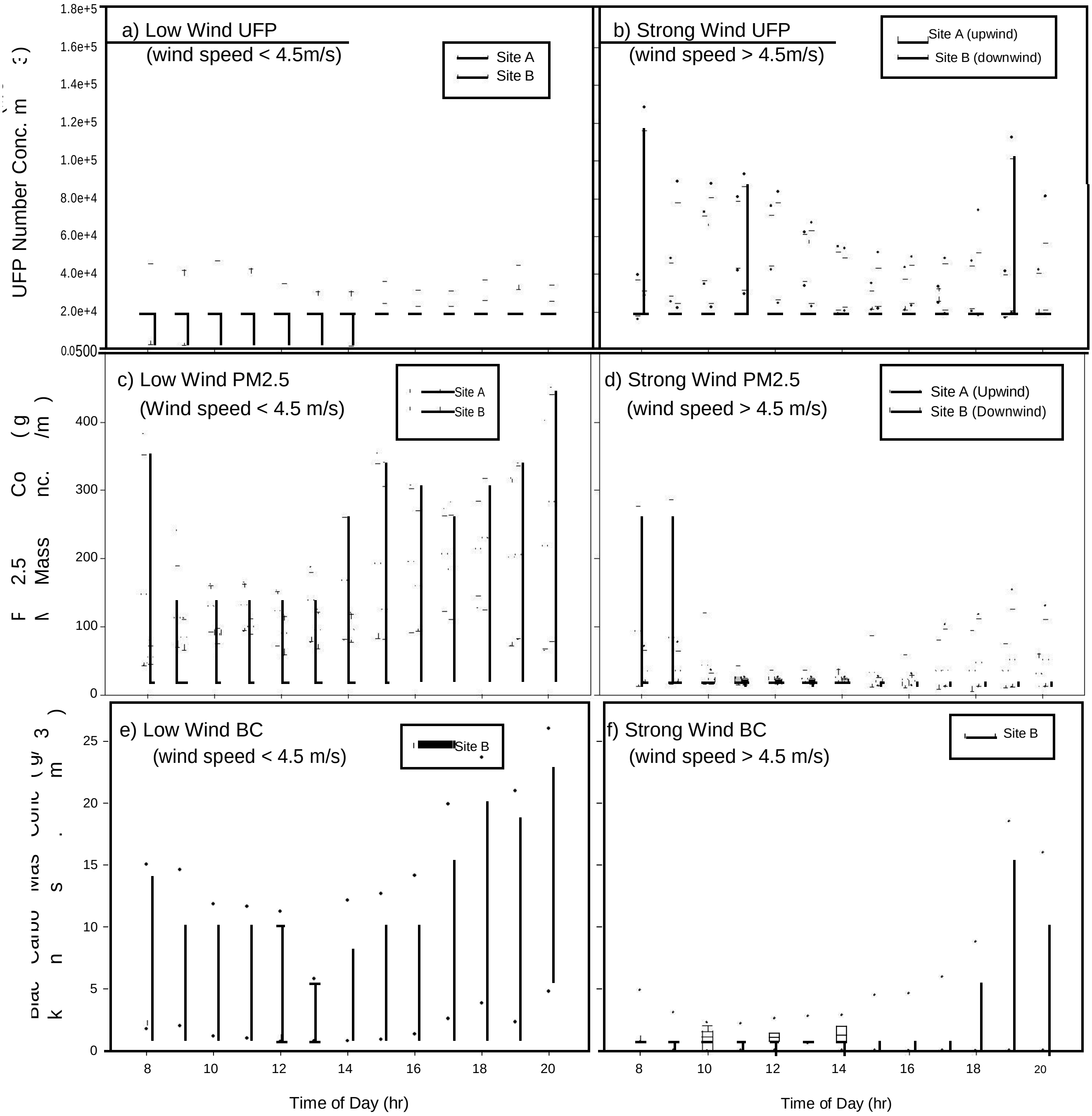

Fig. 5. Diurnal patterns of UFP, PM2.5, and BC concentrations under low wind and strong wind conditions. The lines within boxes are median values. The boundaries of the boxes indicate 25th and 75th percentile of the data, whiskers indicate 10th and 90th percentile of the data, and the points indicate 5th and 95th percentile of the data.

sites used in this study (Shi *et al.*, 2007). Similar temporal profiles were reported in Beijing for hydrocarbon-like organic aerosol (HOA) which were related to cooking emissions (Zhang *et al.*, 2013). Previous studies have shown cooking emissions constitute approximately 20% of organic $PM_{2.5}$ mass in Beijing (Huang *et al.*, 2006; Zhang *et al.*, 2014). At Site A, UFP temporal profile also suggests the influence from Chinese style cooking because the two modes were at approximately lunch and dinner hours, and there are many student dining halls located on campus as illustrated in Fig. 1(c) . Most of the dining halls are located close to sampling Site A. One major dining hall (PKU Dining Hall) is about 100 meter southwest to Site A, and another one (Yannan Food) is about 200 meter west to Site A. There are many other dining halls on campus that are located within 500 meters from Site A. At Site B, UFP concentrations show bimodal at the morning and evening time during the traffic rush hours at early morning and late afternoon (Fig. 5(b)).

The average UFP number concentrations at Site A were similar during the low wind and strong wind time, but the variations were greater on strong wind days (Figs. 5(a) and 5(b)). This indicates that stable atmosphere conditions allowed UFP to coagulate faster, but the strong winds made UFP disperse faster. In addition, under the low wind condition, $PM_{2.5}$ levels are much higher which may provide more surface areas for UFP to deposit (Figs. 5(a) and 5(b)). In contrast to UFP number concentrations, average $PM_{2.5}$ and BC mass concentrations were higher under low wind and had higher variations compared with strong wind. This diurnal pattern is consistent with the log-decay mode in Fig. 4 for the $PM_{2.5}$ variations with wind speeds, especially during the daytime when the inversion effects are not strong and the atmosphere is relatively unstable (Figs. 5(c) and 5(d)). When the wind speeds were high, low variability of $PM_{2.5}$ and BC was observed (Figs. 5(d) and 5(f)). Traffic density is an important factor influencing roadside UFP levels. The HDVs (heavy duty vehicles), including buses and transport trucks but not heavy duty diesel trucks (HDDVs), comprise about 11.8% of the total traffic. The HDV vs. LDV (light duty vehicle) ratio was about 0.13. The traffic pattern on Chengfu Rd. shows that hourly traffic densities may become quite high during the morning and afternoon rush hours (8 AM–9 AM, 4 PM–7 PM, respectively), and is capable of reaching over 2500 vehicles per hour on average.

The overall hourly traffic densities were analyzed and compared with the corresponding hourly UFP concentrations at Site B. The analysis excluded data from periods when Site B was upwind of both Chengfu and N. Zhongguancun Roads. Average hourly UFP was found to have nearly no correlation with hourly total traffic densities ($R^2 = 0.05$). However, there was a relatively higher correlation between hourly UFP and HDV densities with an R^2 of 0.40.

CONCLUSIONS

In this study, traffic related pollutants including UFPs, $PM_{2.5}$, and BC were measured concurrently at a campus and a roadway sampling site near PKU campus from December 10th through 23rd, 2011.The traffic density data were collected with recorded video footage. Observed ambient UFP concentrations, $PM_{2.5}$, and BC mass concentrations all showed high averages and strong variations at both sampling sites. The averages of UFP concentration and $PM_{2.5}$ mass concentration at roadside were not significantly higher than campus site indicating local emission sources on campus. The regression results indicate strong wind speeds influenced $PM_{2.5}$ and BC mass concentrations more than UFP concentrations. Temporal profiles of UFPs showed distinct impacts of traffic on roadway site measurements and Chinese style cooking as a potential source impacting campus site measurements. The high $PM_{2.5}$ concentrations in Beijing might suppress UFP concentrations by providing a condensation sink for semi-volatile vapors and extra surface areas for particle deposition.

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Measuring and modeling air exchange rates inside taxi cabs in Los Angeles, California

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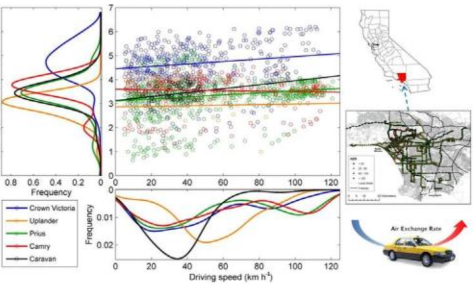
highlights

- Air exchange rates in 22 representative Los Angeles taxi cabs were quantified.
- AERs under realistic driving condition had a mean of 63 h^{-1} and a median of 38 h^{-1}.
- AERs were significantly higher when driving on freeways than on local streets.
- With medium fan speed under out-door air mode, average AERs increased 32%.

graphical abstract



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a b s t r a c t

Air exchange rates (AERs) have a direct impact on traffic-related air pollutant (TRAP) levels inside vehicles. Taxi drivers are occupationally exposed to TRAP on a daily basis, yet there is limited measurement of AERs in taxi cabs. To fill this gap, AERs were quantified in 22 representative Los Angeles taxi cabs including 10 Prius, 5 Crown Victoria, 3 Camry, 3 Caravan, and 1 Uplander under realistic driving (RD) conditions. To further study the impacts of window position and ventilation settings on taxi AERs, additional tests were conducted on 14 taxis with windows closed (WC) and on the other 8 taxis with not only windows closed but also medium fan speed (WC-MFS) under outdoor air mode. Under RD conditions, the AERs in all 22 cabs had a mean of 63 h^{-1} with a median of 38 h^{-1}. Similar AERs were observed under WC condition when compared to those measured under RD condition. Under WC-MFS condition, AERs were significantly increased in all taxi cabs, when compared with those measured under RD condition. A General Estimating Equation (GEE) model was developed and the modeling results showed that vehicle model was a significant factor in determining the AERs in taxi cabs under RD condition. Driving speed and car age were positively associated with AERs but not statistically significant. Overall, AERs measured in taxi cabs were much higher than typical AERs people usually encounter in indoor environments such as homes, offices, and even regular passenger vehicles.

1. Introduction

Previous studies have found that, for the general U.S. population that spends on average 1.3 h driving each day (Klepeis et al., 2001), 17e50% of their total daily ultrafine particles (UFPs)

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exposure is from in-vehicle exposures (Fruin et al., 2008; Wallace and Ott, 2011; Zhu et al., 2007). For taxi drivers, this in-vehicle percent of daily UFP exposure is likely much higher because, on average, they work six days per week and spend 7e12 h each day on driving (LA DOT, 2010). Previous studies have shown that professional drivers have high occupational exposure to traffic-related air pollutant (TRAP) (Gustavsson et al., 2000), which is also reflected by different biomarkers (Brucker et al., 2013; Gustavsson et al., 1996). Therefore, Knibbs and Morawska (2012) pointed out there is a great potential to link the exposure science and epidemiology by studying professional drivers' exposure and illness.

Air exchange rate (AER) is a factor that affects the air quality in a defined space, in this case, the taxi cabin (Chan and Chung, 2003; Knibbs et al., 2010). Higher AER leads to lower concentrations of air pollutants that are originated indoors and higher concentrations of air pollutants that are originated outdoors. In the case of motor vehicles, higher AER would result in higher in-cabin concentrations of TRAPS and lower concentrations of in-cabin-originated compounds such as phthalates (Geiss et al., 2009) and alkanes (You et al., 2007). Therefore the AERs in the cabin would impact the drivers' occupational exposure to different types of air pollutants in different ways. Quantifying the AERs in taxi cabins would lead to better understanding of taxi drivers' occupational exposure to air pollutants from different sources. Several studies have measured the AER in regular passenger vehicles (Fletcher and Saunders, 1994; Knibbs and de Dear, 2010; Knibbs et al., 2010). For example, Ott et al. (2008) measured AERs in four motor vehicles at six different driving speeds ranging from 20 to 72 mph (32e116 km h–^{1}). With windows closed and ventilation system off, the AER was less than 6.6 h–^{1}. They also found that the position of windows, air conditioning (AC) system settings, driving speed, car model, and car age all have significant effects on AER. Fruin et al. (2011) successfully built a statistical model to predict vehicle AER using car age, mileage, manufacturer, and driving speed, based on measurements conducted in 59 passenger vehicles at three different speeds. According to this model, a typical California passenger vehicle manufactured in 2010 would have AER of 20 h– ^{1}, when driving at speed of 105 km h–^{1}. However, these studies were conducted under controlled experimental conditions, so that some of the influencing factors such as driving speed, windows position, and air conditioning settings were arbitrarily selected. Therefore, it is expected that AERs published in the literature may not reflect true AER values for taxis, because these influencing factors are changing frequently under realistic driving conditions.

In addition, the characteristics of taxi cabs are potentially quite different from those of regular passenger vehicles. For example, taxi cabs are likely to have higher mileages than regular passenger vehicle of same age and tend to be leakier because of excessive wear and tear. Consequently the AERs for taxis under realistic driving conditions are not fully understood. Previous studies conducted on passenger vehicles may not sufficiently represent the AERs in taxi cabs (Fletcher and Saunders, 1994; Fruin et al., 2011; Knibbs et al., 2009; Ott et al., 2008).

To fill this data gap, the first objective of this study is to measure the AERs in a number of representative taxi cabs under realistic driving (RD) conditions, to quantify the typical AERs experienced by taxi drivers in the Greater Los Angeles area. The second objective is to investigate if keeping windows closed (WC) and/or using medium fan speed (MFS) would significantly change the AERs in taxi cabs. A GEE model based on measurement data is used to analyze the importance of various factors that influence AERs. The measured AERs and modeling results may be applied to other taxi cabs in the Greater Los Angeles area.

2. Methods

2.1. Taxi cab recruitment

A recruitment/survey campaign was conducted at the Los Angeles Airport (LAX) taxi holding lot from February 11th to 15th, 2013, in order to recruit study participants and collect basic information about taxi drivers and their cabs. A questionnaire that included 10 questions about age, race, smoking history, car model, car age, and driving related behavioral factors was designed and used for the recruitment/survey campaign (see Supplementary Material Fig. S1). A total of 2449 survey forms were handed out and 316 complete survey forms were collected. The descriptive statistics of these 316 survey forms are also provided in the Supplementary Material Table S1. Out of these 316 taxi drivers, 121 non-smokers were eligible to participate in this study. To ensure the sampled taxi drivers/cabs are representative, stratified random sampling was conducted based on car models and drivers' age. Drivers' age was considered as a factor for stratified random sampling because previous studies have found that drivers of different ages have different driving patterns (Horberry et al., 2006), which affect both the in-cabin air exchange rates (Hudda et al., 2011) and the vehicles' pollution emissions (Ericsson, 2001). A total of 22 taxi drivers/taxi cabs out of 121 eligible drivers were selected to participate in this study. The study design and protocol have been approved by the Institutional Review Board (IRB) of University of California Los Angeles. In terms of vehicle models, the 22 taxi cabs included 10 Prius, 5 Crown Victoria, 3 Camry, 3 Caravan, and 1 Uplander.

2.2. Experiment design

Each experiment consisted of four consecutive test days with one driver and his or her taxi cab. On each test day, the driver drove 6 h in the Greater Los Angeles area as he or she would typically do. One field technician rode along in the taxi cab operating and maintaining all the sampling instruments. The starting time of each day was based on the driver's availability and kept consistent for each driver during the four test days in order to minimize the differences in traffic conditions and meteorological conditions among the four test days. No actual fares were collected during the tests and the drivers' time and efforts were compensated by the research funding. Each driver was allowed to take breaks as he or she would during a typical work day. The time and location of each break were recorded by hand and confirmed by a GPS unit (Qstarz GPS BT-1000XT, Taipei, Taiwan). Data collected during the breaks were not used to calculate AERs. The driving routes were not specifically planned for each driver. Instead, on the first test day, each driver was asked to drive from the start location, University of California Los Angeles, to the area where he or she usually works and repeat what he or she did in the previous day. The same route was used as much as possible for the following three test days, to minimize the difference among different days. In total, measurements were conducted on 83 different days from April 2013 to November 2013. Five test days were lost due to two Caravan drivers only partially completed their four-day testing. The total mileage driven by the 22 taxi drivers in this study was approximately 11,000 km and the total hours of field measurement was approximately 500 h.

Three experimental conditions: realistic driving (RD), windows closed (WC), and windows closed with medium fan speed (WC-MFS) were used in this study. Under RD condition, everything was kept as close to the driver's everyday working conditions as possible and the drivers had control over all the vehicle operations such as opening/closing windows, turning air conditioning (AC) on or off, setting ventilation to recirculation or outdoor air mode. The

AERs measured under RD condition best represent the AERs experienced by Los Angeles taxi drivers during their everyday job. Under WC condition, the taxi drivers were required to keep all the windows closed during the 6 h measurement, but the taxi drivers had control over AC and ventilation mode. WC condition was used to investigate whether keeping windows closed all the time could reduce the AER in a taxi cabin. Under WC-MSF condition, the taxi drivers were not only required to keep all the windows closed but also keep the ventilation fan at mid-level under outdoor air mode throughout the 6 h measurement. However, the drivers were allowed to turn the AC on or off to achieve a thermally comfortable environment in the taxi cabin. The operation of AC only affects the working status of the air heating/cooling components and does not change the air flow in the ventilation system, therefore has minimum impacts on AERs. For the first 14 drivers, RD and WC conditions were used, each for 2 days. While for the rest 8 drivers, RD and WC-MFS conditions were used, each for 2 days.

2.3. AER measurements

The AER measurements were conducted by using CO_2 as a tracer gas, a method which has been proven to be relatively easy yet accurate in regular passenger vehicles (Fruin et al., 2011). Two Q-Trak Indoor Air Quality Monitors (TSI Inc., MN USA) were used to measure simultaneously the in-cabin and on-road CO_2 concentrations. One Q-Trak had its probe placed outside, usually attached to the top of taxi cab, to measure the on-road CO_2 concentrations. The other Q-Trak was placed in taxi cab and the probe was fixed at the center of the cabin, between taxi driver and the field technician who sat on the back seat. Previous studies have shown that the well-mixed condition of in-cabin CO_2 can be assumed (Fruin et al., 2011; Hudda et al., 2011; Lee and Zhu, 2014). Both Q-Traks were reading and recording CO_2 concentrations at one second time resolution. Later on these CO_2 concentrations were averaged by one minute to reduce data fluctuation. The two Q-Traks were calibrated using gas standards in the lab and then collocated with each other at different CO_2 levels. The collocation test showed that the readings from the two units correlated well ($R^2 > 0.95$), although not exactly the same. Thus, the readings from the in-cabin unit were corrected against the on-road unit, by using the linear correlation equation obtained from the collocation test, assuming the on-road unit was the 'gold standard'. An AER model based on CO_2 mass balance in the taxi cabin was built, as shown in Equation (1).

$$V\frac{dC_i}{dt} = E + QC_o - QC_i \quad (1)$$

where V is the volume of cabin in m^3, which is reported by the vehicle manufacturer, C_{in}, and C_o are the in-cabin and on-road CO_2 concentrations in mg m^{-3}, respectively, E is the emission rate of CO_2 in the cabin in mg min^{-1}, and Q is the air flow rate from outside into the cabin in m^3 min^{-1}. By rearranging Equation (1) and using its discrete form, Equation (2) was obtained and used to calculate the AER at time t.

$$\frac{C_{in}(t+\Delta t) - C_{in}(t-\Delta t)}{2\Delta t} = AER(t)[C_o(t) - C_{in}(t)] + E/V \quad (2)$$

where AER is a function of time and equals to Q/V in min^{-1}. During the data processing, all data were averaged over one minute, to reduce the noise from the instruments. Therefore the Δt in Equation (2) was one minute and all the AERs calculated in this study were one-minute averaged values and converted in unit of h^{-1}.

The measured GPS coordinates and AERs were matched by the instrument time stamps during data processing. Then a piece of computer code was used to check if the taxi was on freeways or on local streets, by using an identification algorithm based on the GPS coordinates and the Los Angeles GIS database, which was downloaded from the Los Angeles County GIS Data Portal (http://egis3. lacounty.gov/dataportal).

2.4. AERs under WC and WC-MFS conditions

To explore how the operations on taxi cabs' windows and ventilation fan affect AERs for each individual taxi, the geometric means of AERs in individual taxi cabs under different conditions were calculated and then the paired two-sample Wilcoxon test was used for statistical inference.

2.5. GEE model for AERs

A Generalized Estimating Equation (GEE) model was developed to analyze the importance of different factors that can potentially impacts AERs. Many factors, such as driving speed, car age, car mileage, and the driver themselves all have great potential to impact the AER in a taxi cabin during driving (Fruin et al., 2011; Knibbs et al., 2009). As previously found (Fruin et al., 2011) and confirmed in this study, the AERs measured repeatedly on the same vehicle were correlated. For example, an old Ford Crown Victoria will consistently have higher AERs than a new Toyota Prius across all driving speed, given same window position and ventilation settings. Therefore GEE model was selected because it is a widely used statistical model for data collected from repeated measurements on the same statistical units, in this case, the taxi cabs. A series of models were built in R software by using the GEE package. All the aforementioned potential influencing factors were used as input variables and their individual significance in the models was calculated. The model with the smallest Quasi Akaike Information Criterion (QIC) was chosen as the final model. Since the distribution of measured AERs was highly right-skewed, natural log transformation was performed on AERs before fitting the model.

The value of in-cabin CO_2 emission, E, was estimated based on the number of people in the vehicles. During all the experiments, there were two people inside the vehicle during driving: the driver and the field technician. It was assumed that the driver and the technician, at the activity level between sedentary to lightly active, generates 0.68 L CO_2 per minute (McArdle et al., 2010), which equals to one adult generating 900 g of CO_2 in a day. The authors are aware of the uncertainty in this respiration CO_2 production rate. Due to the fact that all the tested drivers are adults with BMI of 26.7 ± 4.5 kg m^{-2}, the uncertainty of E was estimated to be ±16%. Since the calculated AERs were linearly related to the E, the uncertainty in calculated AERs was approximately ±20%, taking other uncertainties, such as the ±10% accuracy of Q-Trak readings, into account.

3. Results and discussion

3.1. Characteristic of taxi cabs

The data collected from the survey questionnaires demonstrated that taxi cabs have higher mileages than a passenger vehicle of the same age. As shown in Fig. 1, a Los Angeles taxi cab has approximately twice as many mileages as a regular California passenger vehicle of the same car age. Therefore the taxi cabs are expected to have more wear and tear, and are likely to be leakier than regular passenger vehicles.

The 22 tested taxi cabs have a similar distribution of car model when compared with the surveyed results from the recruitment campaign. The Toyota Prius and Ford Crown Victoria are the two

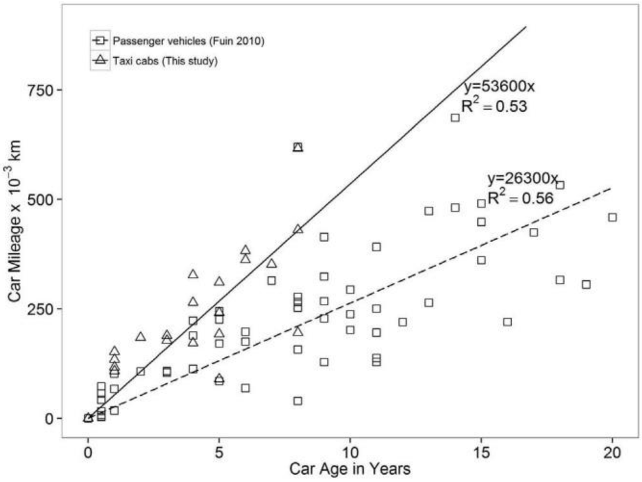

Fig. 1. Relationship between car age and car mileage, comparing taxi cabs and representative passenger vehicles.

most popular car models among Los Angeles taxi cabs. The five car models tested in this study e Prius, Crown Victoria, Camry, Caravan, and Uplander e comprised 86% of all types of taxis in Los Angeles.

For drivers' working pattern and habits of operating windows and ventilation system, the questionnaires showed that the taxi drivers work 6.1 ± 0.8 days per week and 11.9 ± 2.3 h on each working day, and 18% of taxi drivers spend more than 6 h each day driving on freeways. In addition, 56% of taxi drivers keep their windows open for at least half of their work time. However, field technicians observed that, five of the taxi drivers kept the windows closed for roughly 90% of total testing hours even during the RD condition test. It is possible that the taxi drivers' behaviors were affected by the experiment and they closed windows more than they usually do on their job. If this can be confirmed, the AERs measured under RD condition in this study may underestimate the AERs taxi drivers experience on their job.

3.2. Measurement results

3.2.1. CO_2 concentration profiles

Typical one-minute averaged in-cabin and on-road CO_2 con-centrations as well as the driving speed are shown in Fig. 2.

During this period, there are two people inside the taxi with an estimated CO_2 emission rate of 0.68 L per minute. As shown in Fig. 2, the driving speed increased and the in-cabin CO_2 concen-tration decreased around 20:45, but the on-road CO_2 concentra-tions remained relatively stable. This suggests higher driving speed led to higher AER which reduced the in-cabin CO_2 concentrations.

3.2.2. AERs under RD condition

Fig. 3 shows the map of the driving routes covered in this study and the spatial distribution of AERs measured under RD condition. There is no clear spatial clustering pattern of the data, indicating that the location is not an important factor in determining the AERs. The relationship between driving speed and AERs measured under RD condition for all 22 cabs are presented in Fig. 4, which is a scattered plot with the distribution for both axes. In general, taxi driving speeds (the x axis) had two modes corresponding to driving on local streets and freeways, which is typical for urban traffic conditions. The log-transformed AERs (the y axis) also had two modes, one of which had higher value and higher frequency than the other. The descriptive statistics of AERs measured under RD condition are shown in Table 1.

3.2.3. AERS under WC and WC-MFS conditions

Fig. 5 shows the comparison between AERs measured under RD condition with those measured under WC condition and WC-MFS condition. Fig. 5a shows that WC conditions had AERs that are not significantly different with those under RD conditions in this study. This is possibly due to the fact that most drivers kept their windows closed even during their RD condition test, as observed by the field technicians. Previous studies have found that closing windows can reduce AERs in vehicle cabins, when other operation conditions held constant (Esber et al., 2007; Fruin et al., 2011; Ott et al., 2008; Park et al., 1998). It is reasonable to believe that, for those taxi drivers who open their cab windows often on their job, closing windows can potentially shelter them from high AERs and high TRAP concentrations. Compared with the RD condition, the WC-MFS condition significantly increased the AERs in taxi cabs. Based on data collected from the eight taxi cabs (Fig. 5b), the mean AERs under WC-MFS condition, 37 h^{-1}, was 32% higher than those measured on RD condition, which was 28 h^{-1}. This finding high-lighted the importance of improving the cabin filter efficiency to mitigate the taxi drivers' occupational TRAP exposure (Lee and Zhu, 2014).

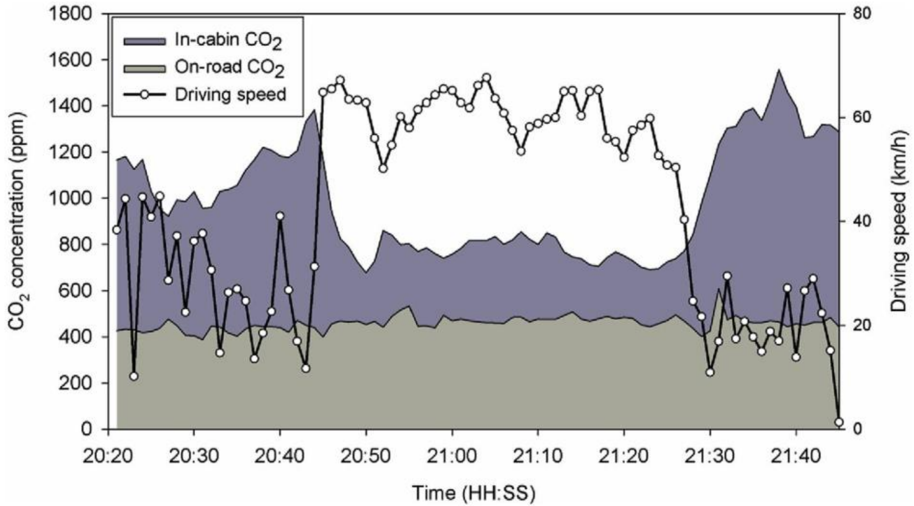

Fig. 2. Typical in-cabin and on-road CO_2 concentrations and driving speeds.

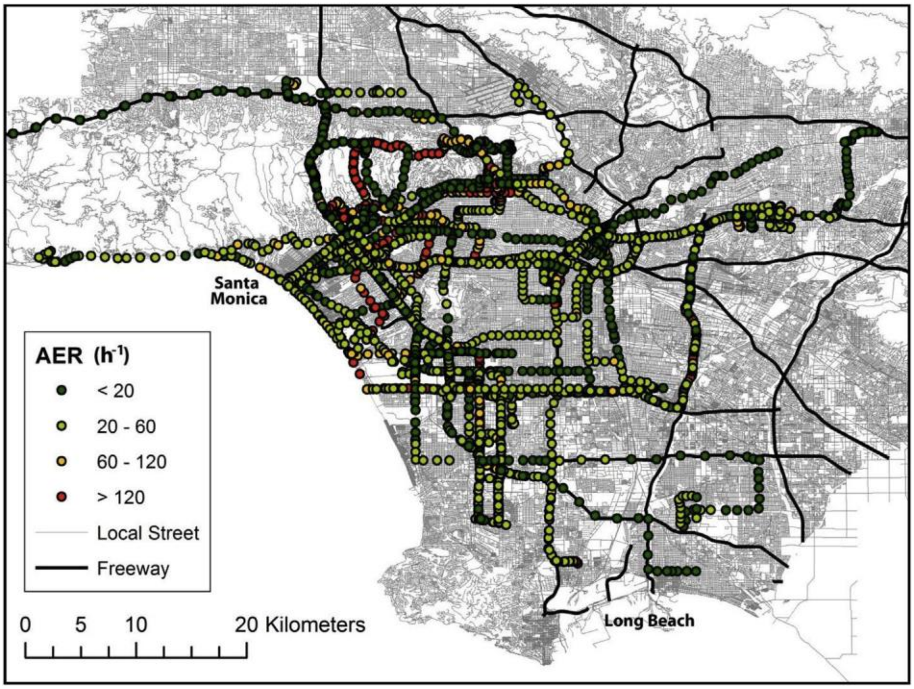

Fig. 3. Map of driving routes and the spatial distribution of AERs measured under RD condition.

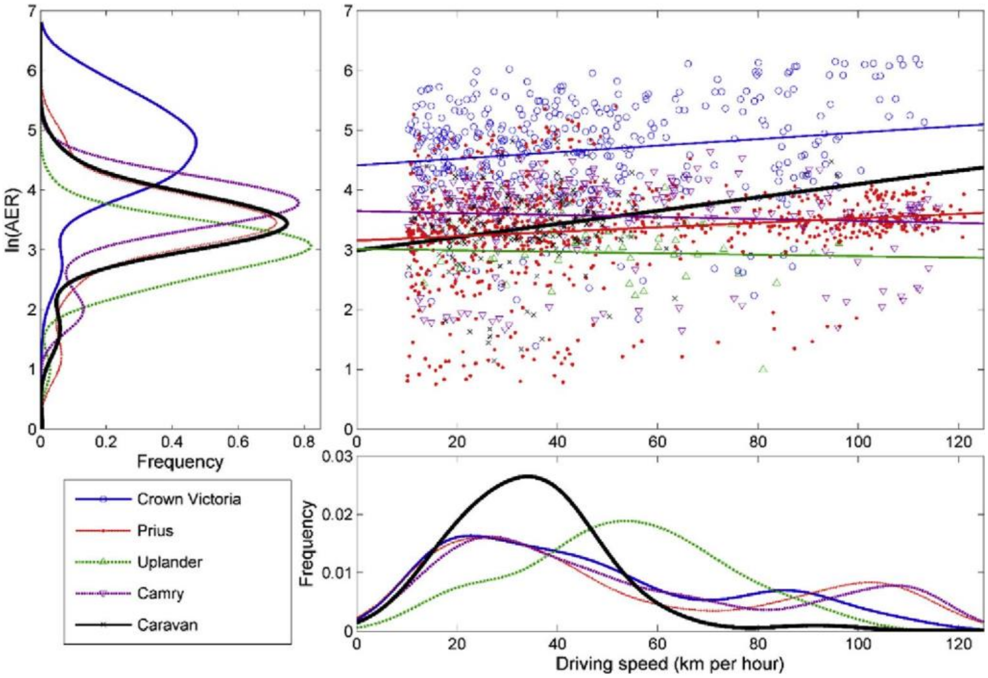

Fig. 4. Log-transformed air exchange rates and its relationship with driving speed, grouped by car models. Air exchange rates were measured under RD condition.

3.2.4. Comparing AERs obtained on local streets and freeways

After log-transformation was performed on AERs, Welch's t-test was used to test if the AERs obtained from freeways and local streets are significantly different. As shown in Fig. 6, the AERs ob-tained on freeways were statistically higher than those obtained on local streets, in all three experimental conditions. This finding suggested that the taxi drivers are experiencing higher exposure to TRAP when driving on freeways, since not only AER but also the air pollutant levels are also usually higher on freewayst han on local streets (Quiros et al., 2013; Shu et al., 2014; Zhu et al., 2007). Therefore, using TRAP mitigation methods in taxi cabs, especially when driving on freeways, would be important to reduce the occupational exposure to TRAP (Lee and Zhu, 2014).

3.3. Modeling results

The GEE model with the lowest QIC, which was selected as the final model, is presented by Equation (3).

Table 1
Summary of characteristics of tested taxi cabs and air exchange rates measured under RD condition.

Car model	Number of vehicles	Mileage [10–^{3} km ± SD]	Car age [Years ± SD]	AER under RD condition [a] [h–^{1}]				
				n [b]	Mean	25%	50%	75%
Camry	3	190 ± 74	2.3 ± 1.5	528	37.0	14.7	41.3	82.3
Caravan	3	283 ± 132	3.7 ± 2.5	502	18.9	13.6	24.0	32.8
Uplander	1	433 ± 0	8.0 ± 0	158	26.8	16.4	28.2	49.4
Crown Victoria	5	446 ± 151	7.0 ± 1.0	1126	75.2	41.3	78.3	148.4
Prius	10	184 ± 63	4.0 ± 2.1	2057	18.9	7.1	25.5	39.6

[a] AER is in unit of h–^{1} before log transformation.
[b] Number of 5-min average AER values.

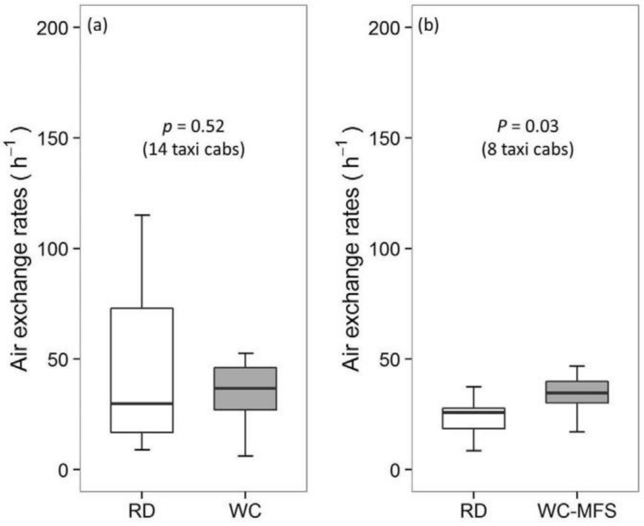

Fig. 5. Air exchange rates measured under RD condition compared with those measured under WC condition and WC-MFS condition, respectively. Each point represents the geometric mean of AERs obtained in one taxi cab under different conditions.

ln(AER) ¼ b_0 þ b_1 _ Speed þ b_2 _ CarAge þ b_3 _ Mileage þ b_4 _ CarMode þ ε_i (3)

where AER is the observed air exchange rates in h–^{1}, Speed is the driving speed in km h–^{1}, CarAge is a continuous parameter which is the number of years between the car model year and 2014, Mileage is the odometer reading converted into 10^{3} km, ε_i is the adjustment for individual taxi cab/driver combination, and CarModel is a categorical parameter that has five levels: Camry, Caravan, Uplander, Crown Victoria, and Prius. Prius was used as the reference category. Independence correlation structure was used in the model for repeated measures. Measurement data obtained under RD condition were fitted in Equation (3) to obtain model parameters. The coefficients are listed in Table 2.

The GEE model provided generalized estimates of linear model, taking the individual-specific effect of each taxi cab/driver combination into consideration. The modeling results showed large intercepts, which indicated that AERs are generally high under RD condition. The Speed, CarAge, and Mileage all had p-value greater than 0.05, meaning they are not statistically significant factors in determining the AERs in taxi cabs. However, the Speed and CarAge were positively correlated with AERs, which is consistent with the finding showing higher AERs when driving on freeways, where the speed was also higher than on the local streets. It is consistent with the findings in previous studies on passenger vehicles (Fruin et al., 2011; Park et al., 1998). The reason that speed was not statistically significant in this study was that the experiment conditions used in previous studies and this study were different. Those previous studies used well-controlled experiment conditions, i.e. closed-window, recirculation ventilation, and stable driving speed, aiming to understand the mechanisms of in-vehicle air exchange and

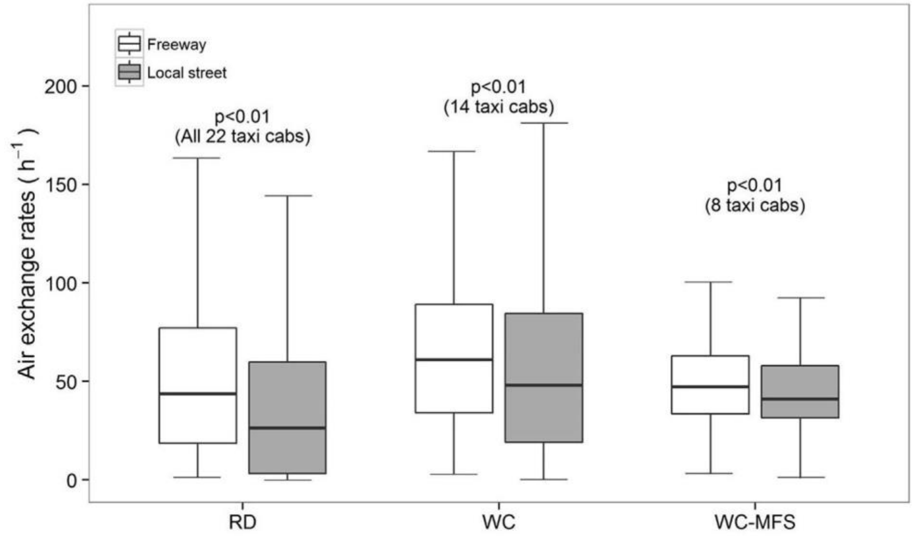

Fig. 6. Comparison between air exchange rates obtained while driving on freeways and on local streets.

Table 2
Coefficients of generalized estimating equations model for AERs measured under RD condition.

	Estimate	Std. Err.	p-Value
Intercept	3.026	0.306	<0.001
Speed	0.003	0.002	0.142
CarAge	0.039	0.069	0.574
Mileage	_0.002	0.002	0.201
CarModel: Prius [a]	0	0	
CarModel: Camry	0.714	0.480	0.137
CarModel: Caravan	0.252	0.320	0.634
CarModel: Uplander	0.793	0.378	0.036
CarModle: Crown Victoria	2.305	0.341	<0.001

[a] Prius was used as the reference category therefore its estimate and standard deviation were both zero.

its determining factors. For example, in the Fruin et al. (2011) study, the AERs inside regular passenger vehicles under “nearly constant driving speed” were treated as a constant value, since other factors such as windows position and ventilation settings were fixed. This study, instead of studying the underlying mechanisms of air ex-change in vehicles, aimed to quantify the actual AERs in taxi cabs during taxi drivers' representative working conditions, under which all the aforementioned factors are frequently changing. That is why the AERs measured in this study had large variability (with a geometric standard deviation of 3.3), therefore the statistical significance of speed was reduced. The highly variable AER data sug-gested that the taxi drivers' occupational exposure to TRAP is also highly variable, even within the same workday.

On the other hand, the modeling results suggested that the AERs measured in two models of taxi cabs, Uplander and Crown Victoria, whose manufacturers were GM and Ford respectively, had signifi-cantly higher AERs than those measured in other three models, two of which were manufactured by Japanese companies. Interestingly, this is also consistent with one of previous studies which demon-strated that passenger vehicles manufactured by GM and Ford also had significantly higher AERs than vehicles manufactured by Jap-anese companies (Fruin et al., 2011). The reasons for this cross-brand different in AERs are outside of the scope of this study and deserve further investigation. The data obtained in this study provided more ‘realistic’ information about the actual AERs the taxi drivers experience in their daily work and can be linked to their occupational exposure assessment in further studies.

3.4. Implications for occupational exposure

Under high AER conditions, on-road air enters the taxi cabin at a higher flow rate, carrying TRAP into the cabin. Because taxi drivers spend substantial amounts of time driving, their occupational exposure to TRAP could be orders of magnitude higher than the general population (Knibbs et al., 2010). Two pollutants of partic-ular concern are UFPs and particle-bound polycyclic aromatic hy-drocarbons (PB-PAH) (Brucker et al., 2013). These two pollutants are generated by the fuel burning process in motor vehicles engine thus have the highest concentrations on the road (Hu et al., 2009; Zhu et al., 2002). Further studies on the in-cabin air pollutants concentrations and in-cabin-to-on-road (I/O) ratios are necessary for more accurate occupational exposure assessment (Hudda et al., 2012). On the other hand, the high AERs could lower the concen-trations of some air pollutants originated inside taxi cabins, for example, phthalates, and thus reduce the taxi drivers' occupational exposure to these chemicals.

Based on the GEE modeling results, switching to taxi cabs of certain models could be an effective way of reducing the AERs experienced by taxi drivers on their daily job, therefore reduce their occupational exposure to TRAP. In fact, this change in taxi cab models is happening in the Greater Los Angeles area, mainly due to the fact that certain vehicle models (i.e. Prius) have higher fuel efficiency than some old models (i.e. Crown Victoria). Thus, a gradual decrease in the Los Angeles taxi drivers' occupational exposure to TRAP can be expected.

Air leakage could be an important source of in-cabin exposure to TRAP. If this is further confirmed, using additional high efficiency air filtering system inside taxi cabs may be a better TRAP exposure mitigation method compared with the existing in-ventilation-duct air filtering method, since a large portion of on-road air are not filtered while entering vehicle cabin (Lee and Zhu, 2014).

3.5. Study limitations

The authors are aware of several limitations of this study. First, more stringent method of determining the CO_2 emission rates of each taxi driver could have been used to increase the accuracy of calculated AERs. To avoid overestimating the AERs, a relatively conservative value of CO_2 emission rate, 0.34 L CO_2 per minute per person was used. Second, the Greater Los Angeles area has a year-round moderate-to-warm weather, which could lead to more frequent window-opening in taxi cabs when compared to areas that have much hotter or colder weather. Therefore cautions should be exercised when extrapolating the AERs data obtained in this study to taxi cabs in other cities.

4. Conclusions

Under the realistic driving (RD) condition, the AERs in the 22 tested Los Angeles taxi cabs had a mean of 63 h^{-1} with a median of 38 h^{-1}. Closing windows did not significantly reduce the AERs in taxi cabs, as the AERs under WC condition were not significantly different from those under RD condition. This is potentially due to the fact, which has been confirmed by the field observation, that most drivers kept their windows closed for most of the time even in the RD condition test. Using WC-MFS condition led to higher AERs in taxi cabs, when compared with those measured under RD con-dition. Driving speed was positively correlated to AERs although not statistically significant. Crown Victoria and Uplander showed significantly higher AERs compared with other three cab models, based on the GEE model results, suggesting switching to models such as Prius could potentially reduce the taxi drivers' occupational exposure to TRAP. In all three experimental conditions, AERs ob-tained on freeways were statistically higher than those obtained on local streets, suggesting driving on freeways can cause higher exposure to TRAP.

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Appendix A. Supplementary data

Supplementary data related to this article can be found at http://dx.doi.org/10.1016/j.atmosenv.2015.10.030.

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Urinary Metabolites of Polycyclic Aromatic Hydrocarbons and the Association with Lipid Peroxidation: A Biomarker-Based Study between Los Angeles and Beijing

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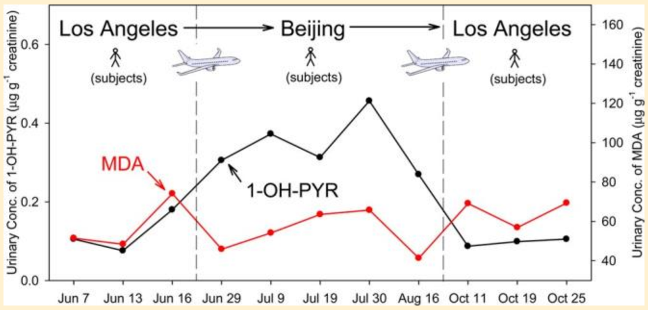
Supporting Information

ABSTRACT: Air pollution is among the top threats to human health in China. As air toxicants, polycyclic aromatic hydrocarbons (PAHs) could bring significant risks to population; however, the exposure to PAHs in China and its health impact are not fully understood. In 2012, a summer exchange program allowed 10 students to travel from Los Angeles to Beijing and stay there for 10 weeks. Based on the program, this study investigated the difference in urinary concentration of 12 hydroxylated-PAHs (Σ_{12}OH-PAHs) and malondialdehyde (MDA) between the two cities. The median concentration of Σ_{12}OH-PAHs in Beijing (14.1 µg g^{-1} creatinine) was significantly higher than that in Los Angeles (5.78 µg g^{-1} creatinine), indicating a higher exposure in Beijing. The ratios of homogeneous OH-PAHs (e.g., 1-/2-OH-NAP) changed significantly between the two cities ($p < 0.01$), which might suggest a potential alteration in metabolism



INTRODUCTION

Polycyclic aromatic hydrocarbons (PAHs) are a group of air pollutants that contain two or more fused aromatic rings. Their ubiquitous occurrence in the environment has raised increasing concerns due to their high emissions and significant toxicity. The global emission of PAHs was approximately 504 Gg in 2007, of which 21% was from China.[1] PAHs are mainly emitted from combustion sources, such as vehicle emissions, household fuel consumption, and tobacco smoke.[2] All of these sources are geographically close to densely populated areas and could therefore bring significant exposure and health risks. Humans are exposed to PAHs through various pathways including inhalation, ingestion, and dermal absorption.[2−4] For the assessment of the total exposure to PAHs from different routes, urinary hydroxylated PAHs (OH-PAHs), the metabo- lites of PAHs, are widely measured.[2]

PAHs are associated with various adverse health effects (e.g., micronuclei frequency, DNA damage, lung function, and heart could be generated during the metabolism of PAHs. Then, ROS could attack biological molecules such as DNA, proteins, and lipids, resulting in a series of health problems.[6] Malondialdehyde (MDA) is a product of lipid oxidative damage and was widely used as a biomarker for lipid peroxidation.[14,15] MDA was previously found to be associated with both PAHs exposure and various diseases,[14−16] suggesting a potential role of lipid peroxidation between PAHs and the health effects.

In recent years, the severe air pollution in Beijing has created great concerns.[17] As toxic air pollutants, PAHs were also present in higher concentrations in Beijing than in other cities in the developed countries.[18−20] In 2012, the University of California, Los Angeles (UCLA) and Peking University (PKU) carried out a summer exchange program in which a panel of 10 UCLA students traveled to Beijing and stayed for 10 weeks, providing an opportunity to study their PAH exposures and related lipid peroxidation. As shown in a previous study, rate variability),[5−8] and certain adverse health outcomes (e.g., lung cancer, cardiovascular diseases, birth defects, and diabetes).[9−12] The biological mechanism of these associations is not yet clear, and oxidative damage is suggested as a possible cause.[3,13] It has been shown that reactive oxygen species (ROS) repeated measurements on travelers could allow researchers to focus on the impacts of exposure with less interference from individual differences.[21] In this study, the first-morning urine samples of these students were collected before, during, and after the exchange program. A total of 12 urinary OH-PAHs and malondialdehyde (MDA) were measured as surrogates for exposure and lipid peroxidation, respectively. The aims of this study were as follows: (1) assess the exposure to PAHs in Beijing and Los Angeles; (2) characterize the differences in the ratios of OH-PAHs in the two cities to better understand their metabolism; and (3) investigate the association between OH- PAHs andMDA.

MATERIALS AND METHODS

Sample Collection.

All 10 subjects (four males and six females) in this study were healthy UCLA students. The age and body mass index (BMI) of the subjects at the time of sample collection were 23.3 ± 5.8 (mean ± standard deviation; range: 20−39, same as below) years and 21.1 ± 1.4 (18.6− 23.4) kg m^{-2}, respectively. All the subjects were self-reported nonsmokers and participated in the summer exchange program between UCLA (in Los Angeles) and PKU (in Beijing) in 2012. A total of 11 urine samples were collected for each subject before, during, and after the exchange program, with the specific dates shown in Figure 1. Briefly, three urine samples were collected before the program (LA1, from June 7 to June 19) in Los Angeles. A total of five collections were conducted during the program (BJ, from June 29 to August 8) in Beijing. The last three samples were collected after the program (LA2, from October 8 to October 26) when the students returned to the Los Angeles. Because PAHs are metabolized rapidly in animals and human, with half-lives of less than 1 day,[22−24] the urine collection began at least 1 week after arrival in the new city to exclude the interference of previous PAH exposures in the former city. For each urine collection, the first morning urine after fasting for at least 8 h was collected in polypropylene tubes and frozen at −20 °C until analysis.

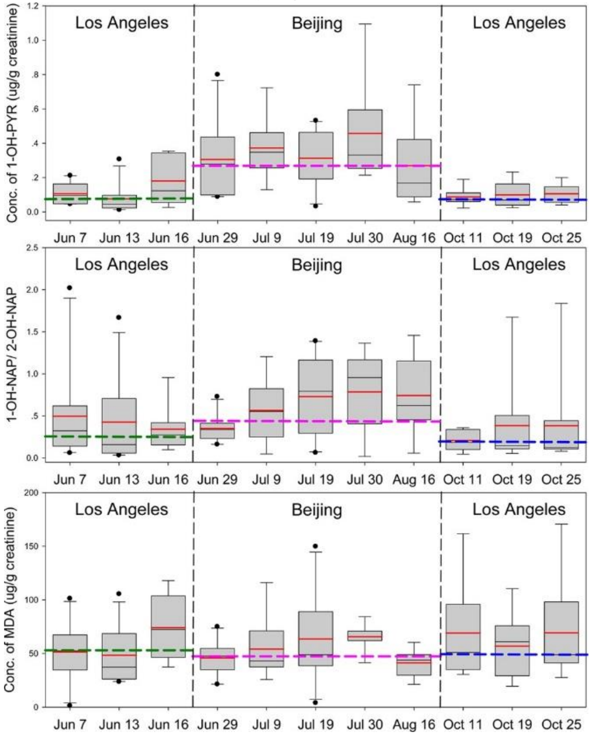


Figure 1. Temporal trend of 1-OH-PYR, 1-/2-OH-NAP, and MDA in urines. Black line, median of each date; red line, mean of each date; green dashed line, median of Los Angeles before the travel (LA$_1$); pink dashed line, median of Beijing; blue dashed line, median of Los Angeles after the travel (LA$_2$); box, 25th and 75th percentiles; whiskers, 10th and 90th percentiles).

For each subject, a questionnaire was used to collect additional information for the 3 days prior to the sample collection. In the questionnaire, detailed information on cooking behaviors (cooking frequency, cooking fuel, and exposure to barbecuing), diet (the consumption of barbecue or baked meat), traffic-related activities (driving hours, public transportation usage, and duration of stay near heavy traffic areas), and secondhand smoke exposure were collected. This study was performed in accordance with the guidelines and approval of the Institutional Review Boards of both UCLA and PKU, and informed consent was obtained from each subject.

Analytical Method.

A previously established method was used in this study to measure the urinary OH-PAHs.[14] Briefly, 2 mL of urine from each sample was spiked with ^{13}C-labeled 3- hydroxyphenanthrene (^{13}C-3-OH-PHE) as surrogate standards and adjusted to pH 5.5 with sodium acetate buffer. Next, the sample was added to 20 μL of β-glucuronidase−sulfatase (*Helix pomatia*, Sigma-Aldrich, St. Louis, MO) and incubated at 37 °C overnight to hydrolyze conjugated phenols. The liquid−liquid extraction with hexane methyl *tert*-butyl ether mixture (9:1, v/

v) was performed three times to extract the analytes. After blowing with nitrogen to near-complete dryness, 0.1 mL of methanol and 1 mL of diazomethane solution were added to the extract, and the OH-PAHs were methylated at room temperature for 5 h. Next, the sample was cleaned with silica gel chromatography (0.6 cm i.d., 6 cm length, with 0.5 cm of anhydrous Na_2SO_4 on top) and eluted with 8 mL of hexane, 8 mL of hexane dichloromethane mixture (3:2, v/v), and 8 mL of dichloromethane sequentially. The analytes were in the second and third fractions. Finally, the sample was concentrated, spiked with d_{10}-acenaphthene (d_{10}-ACE) and d_{10}-phenanthrene (d_{10}- PHE) as internal standards and analyzed using a gas chromatograph and mass spectrometer (GC-MS; Agilent 7890A-5975C) with an electron ionization (EI) ion source and a 30 m DB-5MS column (250 μm i.d., 0.25 μm film thickness; J & W Scientific, Folsom, CA). The monitored ion couples for all analytes and the method detection limits (MDL) (ranged from 7.5 to 18.2 pg mL^{-1}) are listed in Table S1.

Urinary MDA concentrations were measured based on the reaction with 2-thiobarbituric acid (TBA). Briefly, a 150 μL urine sample mixed with 450 μL of TBA and 900 μL of phosphate (0.5 mol L^{-1}) was incubated in water at 95 °C for 1 h. After being cooled and filtered, the mixture was injected into a high-performance liquid chromatograph (HPLC; Waters 2695) with a reverse-phase C18 column (150 mm in length, 3.9 mm i.d.) and a mobile phase of potassium phosphate (0.05 mol L^{-1}, pH = 6.5) and methanol (60:40, v/v). The MDA-TBA adducts could be detected under a wavelength of 532 nm in a UV detector. The detect limit of the method is 7.2 ng mL^{-1}. Urinary creatinine was measured by a spectrometer under a wavelength of 510 nm based on the Jaffe reaction.

Quality Control. For each batch of eight urine samples, one laboratory blank sample (with 2 mL of purified water) was

Table 1. Descriptive Statistics of Biomarkers in Urine Samples in Beijing (BJ) and Los Angeles Before (LA_1) and After the Trip (LA_2)

	median (IQR[a])			*p* value[b]			
biomarker	in LA_1 ($n = 30$)	in BJ ($n = 47$)	in LA_2 ($n = 27$)	LA_1 vs BJ	LA_2 vs BJ	LA_1 vs LA_2	Beijing/LA concentration ratio (95%CI; *p* value)[c]
exposure biomarker (ug/g creatinine)							
1-hydroxynaphthalene	0.91 (0.35, 1.45)	1.91 (1.01, 2.62)	0.41 (0.29, 1.05)	< 0.001	< 0.001	0.08	2.6 (1.7−4.0; < 0.001)
2-hydroxynaphthalene	2.58 (1.13, 4.57)	2.74 (1.77, 5.00)	2.43 (1.36, 3.87)	0.34	0.28	0.95	1.1 (0.79−1.5; 0.67)
∑hydroxynaphthalenes	3.08 (1.85, 5.82)	5.01 (2.95, 7.94)	2.76 (1.87, 4.41)	< 0.05	< 0.05	0.57	1.3 (0.97−1.8; 0.08)
2-hydroxybiphenyl	0.45 (0.25, 1.16)	0.45 (0.24, 1.10)	0.66 (0.42, 1.46)	0.87	0.19	0.20	1.1 (0.66−1.7; 0.82)
4-hydroxybiphenyl	0.30 (0.17, 0.45)	1.29 (0.85, 1.98)	0.30 (0.18, 0.43)	< 0.001	< 0.001	0.87	3.7 (2.7−5.0; < 0.001)
4,4′-dihydroxybiphenyl	0.29 (0.18, 0.52)	0.91 (0.76, 1.29)	0.29 (0.18, 0.45)	< 0.001	< 0.001	0.89	2.7 (2.0−3.7; < 0.001)
∑hydroxybiphenyls	1.39 (0.72, 2.22)	2.93 (1.84, 4.79)	1.54 (0.94, 2.77)	< 0.001	< 0.01	0.33	2.3 (1.6−3.2; < 0.001)
2-hydroxydibenzofuran	0.25 (0.14, 0.47)	1.80 (1.24, 2.07)	0.25 (0.20, 0.34)	< 0.001	< 0.001	0.60	6.1 (4.6−7.9; < 0.001)
2-hydroxyfluorene	0.20 (0.10, 0.33)	1.21 (0.80, 1.64)	0.19 (0.12, 0.25)	< 0.001	< 0.001	0.99	5.7 (4.4−7.6; < 0.001)
3-hydroxyfluorene	0.08 (0.04, 0.14)	0.41 (0.31, 0.55)	0.07 (0.04, 0.11)	< 0.001	< 0.001	0.59	5.5 (4.0−7.5; < 0.001)
∑hydroxyfluorenes	0.29 (0.13, 0.49)	1.58 (1.17, 2.31)	0.26 (0.17, 0.34)	< 0.001	< 0.001	0.91	5.6 (4.3−7.4; < 0.001)
1-hydroxyphenanthrene	0.12 (0.06, 0.21)	0.41 (0.30, 0.73)	0.08 (0.06, 0.17)	< 0.001	< 0.001	0.29	3.9 (2.9−5.2; < 0.001)
2-hydroxyphenanthrene	0.08 (0.04, 0.13)	0.26 (0.15, 0.36)	0.06 (0.04, 0.12)	< 0.001	< 0.001	0.53	3.5 (2.7−4.6; < 0.001)
4-hydroxyphenanthrene	0.04 (0.02, 0.07)	0.12 (0.07, 0.20)	0.04 (0.02, 0.10)	< 0.001	< 0.001	0.87	2.7 (1.9−3.9; < 0.001)
∑hydroxyphenanthrenes	0.25 (0.15, 0.43)	0.92 (0.52, 1.26)	0.20 (0.13, 0.36)	< 0.001	< 0.001	0.48	3.5 (2.6−4.6; < 0.001)
1-hydroxypyrene	0.09 (0.05, 0.16)	0.32 (0.18, 0.46)	0.07 (0.05, 0.13)	< 0.001	< 0.001	0.99	3.3 (2.4−4.6; < 0.001)
$\sum_8$hydroxylated PAHs[d]	3.76 (2.16, 7.06)	8.85 (4.99, 12.1)	3.27 (2.37, 5.54)	< 0.01	< 0.001	0.62	1.8 (1.3−2.4; < 0.001)
$\sum_{12}$hydroxylated PAHs[e]	5.77 (3.63, 10.6)	14.1 (7.68, 20.5)	5.78 (3.70, 10.5)	< 0.001	< 0.001	0.85	2.0 (1.5−2.7; < 0.001)
metabolite ratio							
1/2-hydroxynaphthalene	0.29 (0.13, 0.55)	0.53 (0.34, 0.96)	0.15 (0.11, 0.34)	< 0.01	< 0.001	0.14	2.5 (1.7−3.5; < 0.001)
1 + 2/4 -hydroxyphenanthrene	3.99 (2.88, 7.48)	5.08 (4.15, 8.15)	4.00 (2.91, 5.83)	0.07	< 0.01	0.46	1.4 (1.1−1.7; < 0.01)
effect biomarker (ug/g creatinine)							
malondialdehyde	53.7(36.6, 72.5)	48.4 (39.4, 68.3)	49.6 (38.4, 90.8)	0.75	0.40	0.78	0.93 (0.77−1.2; 0.46)

[a] IQR: interquartile range. [b] Mann−Whitney test. [c] Ratio $=10^{\beta}$, where β is the estimated slope for city in multivariate linear regression models with the enter approach and OH-PAHs is log-transformed. Ratios are adjusted by age, gender, and BMI. [d] Sum of hydroxynaphthalenes, hydroxyfluorenes, hydroxyphenanthrenes, and 1-hydroxypyrene. [e] Sum of hydroxynaphthalenes, hydroxybiphenyls, 2-hydroxydibenzofuran, hydroxyfluorenes, hydroxyphenanthrenes, and 1-hydroxypyrene.

prepared. The analysis for blank samples was the same as that for urine samples. For all urine samples, three identical samples were prepared to ensure repeatability. The concentrations of 3- hydroxybiphenyl (3-OH-BP, 15.8%), 2,2′-dihydroxybiphenyl (2,2′-DOH-BP, 12.2%), 3,4′-DOH-BP(14.5%), and 3-hydrox-yphenanthrene (3-OH-PHE, 33.9%) in the blank samples were more than 10.0% of the average concentrations in the urine samples and hence removed from the subsequent discussion. The concentrations of the remaining 12 analytes in blank samples were 1.11 ± 1.05% (average ± standard deviation) of

the average concentrations in urine samples. Thus, blank subtraction was not performed for all those analytes. The relative deviation of all analytes was 21.0 ± 7.2%. The recovery of ^{13}C-3-OH-PHE was 93.6 ± 12.4%. All the OH-PAHs and MDA data were normalized by creatinine.

Statistical Analysis. The Shapiro−Wilk test was applied to check the normality of the data in this study. Median values (with interquartile range, IQR) were reported for urinary biomarkers and their ratios unless otherwise noted. For analytes not detected in urine samples, the 1/2 MDL was applied as a substitute for the statistical analysis. The Mann−Whitney U- test was used to investigate the difference in urinary biomarkers and questionnaire data between the two cities. A two-tailed *p* value of <0.05 was considered significant. Multivariate linear regressions with stepwise or enter approaches were applied to identify the confounding factors and calculate the concentration ratios between the two cities. A simple linear regression model and three linear mixed-effects models were used to investigate the association between MDA and OH-PAHs. In the simple linear regression model (Model A), the association between OH-PAHs and MDA was considered constant among subjects in the two cities:

$$y_{ijk} = \alpha + \beta x_{ijk} + \varepsilon_{ijk} \quad (1)$$

where y_{ijk} and x_{ijk} are the log-transformed concentrations of MDA and OH-PAHs of subject i at time *j* in the city k; respectively. α and β is the fixed intercept and slope, respectively. ε_{ijk} is the residual.

In the three mixed-effects model, a random intercept was allowed among subjects (Model B, eq 2), cities (Model C, eq 3), and both subjects and cities (Model D, eq 4), respectively.

$$y_{ijk} = \alpha + \mu_i + \beta x_{ijk} + \varepsilon_{ijk} \quad (2)$$

$$y_{ijk} = \alpha + \mu_k + \beta x_{ijk} + \varepsilon_{ijk} \quad (3)$$

$$y_{ijk} = \alpha + \mu_i + \mu_k + \beta x_{ijk} + \varepsilon_{ijk} \quad (4)$$

where μ_i and μ_k is the random intercept for subject i and city k, respectively. All statistical analyses were conducted in SPSS package 18.0 (SPSS, Chicago, IL).

RESULTS AND DISCUSSION

Concentrations of Urinary OH-PAHs. For the 12 metabolites of PAHs in the subsequent discussion, the detection rates were all greater than 88%. The concentrationsof OH-PAHs with different numbers of rings are shown in Figure S1. The median concentrations of hydroxynaphthalenes (ΣOH-NAPs, sum of 1- and 2-OH-NAP), ΣOH-BPs (sum of 2-, 4-OH-BP and 4,4′-DOH-BP), 2-hydroxydibenzofuran (2- OH-DBF), ΣOH-FLUs (sum of 2-, and 3-OH-FLU), ΣOH-PHEs (sum of 1-, 2-, and 4- OH-PHE), and 1-OH-PYR were 4.01, 2.12, 0.60, 0.56, 0.43, and 0.13 μg g^{-1} creatinine, respectively. A decreasing trend in urinary concentration of OH-PAHs was observed when the number of aromatic rings increased. This was likely because PAHs with fewer aromatic rings tend to present in a higher concentration in the environment and have a higher urine-excretion rate in human body.[2,22,23,25,26] The concentration of urinary OH-PAHs was influenced by many factors, such as the environmental levels of PAHs, individual physical activities, and individual characteristics. In this study, the determinants of OH-PAHs were investigated using a multivariate model with stepwise approach based on the questionnaire data, and the results are shown in Table S2. The city (i.e., Los Angeles and Beijing) was the dominant factor determining the urinary OH-PAHs concentrations. Individual characteristics (i.e., gender, age and BMI) were also significant factors ($p < 0.05$); however, their impacts on the change of OH-PAHs between the two cities were minimized as multiple measurements were conducted for each subject who serves as his or her own control. The individual physical activities, including diet habits and traffic-related activities, differed significantly between the two cities ($p < 0.05$, Table S3). However, they had limited impacts on the urinary OH-PAHs concentrations in this study as most of them were not significantly associated with OH-PAHs after adjustment for city (Table S2) and thus not considered in the subsequent discussion. Table 1 shows the difference in the creatinine adjusted OH- PAHs concentration between Los Angles and Beijing (unadjusted data are shown in Table S4). It should be noted that biphenyl and dibenzofuran are technically not PAHs but have similar structure and environment sources with PAHs. Hence in addition to the total concentration of all analytes (Σ_{12}OH-PAHs), the total concentration of metabolites of naphthalene, fluorene, phenanthrene, and pyrene (Σ_8OH- PAHs) was also calculated (Table 1). The median concen- tration of Σ_{12}OH-PAHs in Beijing was 14.1 μg g^{-1} creatinine, which was significantly higher than that in LA$_1$ (5.77 μg g^{-1} creatinine, $p < 0.001$) and LA$_2$ (5.78 μg g^{-1} creatinine, $p < 0.001$). No significant difference was observed between the Σ_{12}OH-PAHs concentration in LA$_1$ and LA$_2$ ($p = 0.85$). A similar trend was also observed for ΣOH-NAPs, ΣOH-BPs, 2- OH-DBF, ΣOH-FLUs, ΣOH-PHEs, and 1-OH-PYR. These results indicate that the observed urinary OH-PAHs levels were mainly driven by the differences of various environmental and activity factors between the two cities. On the basis of these biomarkers, it was estimated that the exposure to different PAHs was 1.3−6.1-fold higher in Beijing than in Los Angeles during the study season (Table 1). As a classic biomarker for PAHs exposure, 1-OH-PYR is widely measured in populations around the world.[27,28] Hence, it was used for comparison with other studies. As shown in Figure S2, the concentration of 1-OH-PYR in Beijing (median,0.32 μg g^{-1} creatinine) was higher than that of most cities in developed countries, such as San Francisco, United States (0.08 μg g^{-1} creatinine)[22] and Christchurch, New Zealand (0.04 μg g^{-1} creatinine),[29] but lower than that of most cities in developing countries, such as Nanjing, China (1.08 μg g^{-1} creatinine)[30] and Bangkok, Thailand (0.39 μg g^{-1} creatinine).[31] The concentration of 1-OH-PYR in Los Angeles (0.08 μg g^{-1} creatinine) was comparable to that in cities in developed countries. Those comparisons indicated that the exposure to PAHs in the summer in both Beijing and Los Angeles was at an intermediate level worldwide.

Difference in the Ratios of OH-PAH Isomers. As discussed above, ΣOH-NAPs and ΣOH-BPs differed significantly in the two cities; however, not all metabolites from the same precursor PAHs showed the same concentration ratios between the two cities (Table 1). Briefly, the concentration ratios of 1-OH-NAP, 4-OH-BP, and 4,4′-DOH-BP between Beijing and Los Angeles were significantly greater than 1.0 ($p < 0.001$). In contrast, the concentration ratios of 2-OH-NAP and 2-OH-BP were not significantly different with 1.0. The difference of these OH-PAH isomers indicated potential bias may exist if only one or few isomers were used as surrogates for total PAHs exposures. Instead, the sum of OH-PAHs isomers (i.e., ΣOH-NAPs and ΣOH-BPs) could be the least-biased surrogate for PAHs exposure given the concentrations of multiple OH-PAHs are available.

The reason for the different concentration ratios of OH-PAH isomers between the two cities is unclear, and the interaction between PAHs and cytochrome P450 (CYP) enzymes may be a possible mechanism. PAHs could be metabolized by a series of CYP enzymes, such as CYP1A1 and CYP1B1, through an arene oxide intermediate to form hydroxylated metabolites.[32] Different CYPs in the phase I metabolism of PAHs could result in different metabolite (i.e., OH-PAHs) ratios.[33−35] Meanwhile, PAHs and their metabolites could in turn induce or inhibit the expression of CYPs, which could alter the profiles of CYP enzymes involved in the metabolism of PAHs and then further alter the ratios of OH-PAHs isomers.[33,36] In this study, a higher exposure to PAHs was observed in Beijing, which could possibly cause a shift in the relative expression of different CYPs and might therefore lead to a corresponding shift in the OH-PAHs isomer ratios.

Because previous studies revealed a difference in PAHs metabolite ratios under the catalysis of different CYPs,[33−35] we suspect there may be a link between the alteration of OH-PAHs isomer ratios and the exposure-induced alteration of CYPs expression. To test this hypothesis, we investigated the difference in several OH-PAHs isomer ratios between the two cities. First, the ratio of 1-OH-NAP to 2-OH-NAP (1-/2- OH-NAP) was investigated because (1) 1-OH-NAP and 2-OH- NAP were the only monohydroxylated metabolites of naphthalene so that the ratio would not be influenced by other monohydroxylated metabolites; and (2) the 1-/2-OH- NAP was mathematically independent from the ΣOH-NAPs. As expected, the 1-/2-OH-NAP ratio was significantly elevated in Beijing, suggesting a possible shift in the relative expression of CYPs. It should be noted that the elevation of 1-/2-OH-NAP occurred gradually after the students arrived in Beijing (Figure 1), possibly suggesting the alteration of metabolism could be a subacute process. This may explain the observation in other studies that the variation of OH-PAHs isomer concentrations tended to be more consistent after an accidental high exposure.[23,37]

It should be noted that 1-OH-NAP is also a metabolite of carbaryl pesticides.[38] If 1-/2-OH-NAP was influenced by carbaryl pesticides exposure, we would expect that 1-/2-OH- NAP had a more significant association with ΣOH-NAPs than

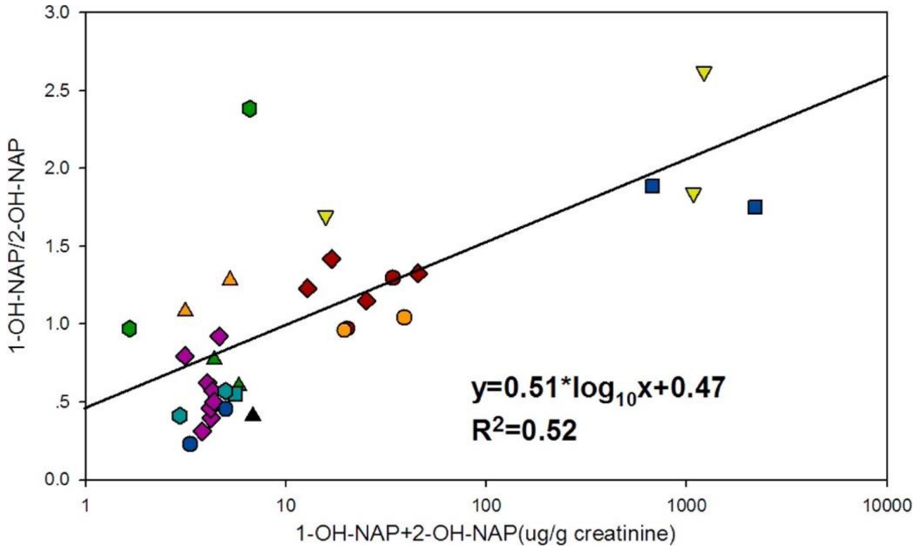


Figure 2. The association between naphthalene exposure and 1-/2-OH-NAP ratio. Red circle, cooking women;[41] orange circle, cooking women;[40] light green downward-facing triangle, coking workers;[52] dark green upward-facing triangle, road construction workers;[43] teal square, healthy general people;[30] dark blue square, coking workers;[53] purple diamond, general people near an aluminum plant;[39] red diamond, coking workers;[6] orange upward-facing triangle, general people near a creosote impregnation plant;[44] black upward-facing triangle, schoolchildren near a road;[54] dark green hexagon, brick kiln workers;[55] teal hexagon, U.S. air forces personnel;[42] blue hexagon, this study. The detailed information on these studies is shown in Table S6.

with other OH-PAHs. However, as shown in Table S5, 1-/2- OH-NAP was not correlated with ΣOH-NAPs but significantly correlated with other OH-PAHs, indicating carbaryl pesticides have limited impacts in this study.

To further confirm the relationship between the 1-/2-OH-NAP ratio and PAHs exposure, we conducted an analysis on selected literature. The selection criteria includes: (1) the population was under a well-defined long-term exposure; (2) the PAHs to which the population was exposed were mainly from combustion sources to minimize the interference from carbaryl pesticide; and (3) the sample size is larger than 10 to decrease the uncertainty caused by individual difference. Because the number and species of the OH-PAHs measured varied among different studies, the concentration of ΣOH- NAPs was used as an indicator for total exposure to PAHs. The results and the description of the literature searches are shown in Figure 2 and Table S6. A significant association was observed between the 1-/2-OH-NAP and ΣOH-NAPs ($R^2 = 0.52$, $p < 0.001$). Additionally, for studies in which repeated measure- ments were conducted that minimized the genetic factors, an interstudy relationship between 1-/2-OH-NAP and ΣOH-NAPs was also observed.[39−44] These results revealed a potential shift in the relative expression of CYPs that might be related to PAH exposures.

The alteration of 1-/2-OH-NAP could explain why 1-OH-NAP and 2-OH-NAP had different concentration ratios between the two cities (Table 1). For 1-OH-NAP, the exposure to PAHs and the corresponding alteration of 1-/2-OH-NAP were in the same direction; therefore, the concentration of 1- OH-NAP was significantly higher in Beijing. However, for 2- OH-NAP, the change in exposure to PAHs could be offset by alterations in the ratio; thus, the concentration of 2-OH-NAP was observed to be similar in the two cities.

This mechanism could also explain the observation of OH- PHEs isomers. Previous studies have shown that 1-OH-PHE and 2-OH-PHE are mainly derived from the same CYPs (e.g., CYP1A1), while 3-OH-PHE and 4-OH-PHE come from other CYPs (e.g., CYP1A2).[22,34,45] These findings were consistent with the observations in this study that the 1 + 2-/4-OH-PHE ratio was significantly elevated in Beijing ($p < 0.01$, Table 1). In addition, 1 + 2-/4-OH-PHE was significantly correlated with several OH-PAHs (i.e., 2-OH-DBF and ΣOH-FLU, $p < 0.05$, Table S5), possibly suggesting a similar link between exposure and metabolism.

Previous studies found that smoking could decrease 1 + 2-/3+ 4-OH-PHE,[22] suggesting exposure to secondhand smoke (SHS) may reduce the 1 + 2-/4-OH-PHE. In our study, SHS exposure is significantly higher in Beijing (Table S3). To distinguish the impacts of PAHs exposure from SHS and non- SHS sources, we divided the data in Beijing into two groups. As shown in Figure S3, all subjects in Beijing had significantly higher ΣOH-PHEs and 1 + 2-/4-OH-PHE than in Los Angeles. Subjects in Beijing with SHS exposures tend to have slightly higher ΣOH-PHEs but lower 1 + 2-/4-OH-PHE compared with those without SHS exposures, which was consistent with previous studies on smoking.[22] However, no significant difference was observed between subjects with and without SHS exposures in Beijing. These results indicate that the elevation of ΣOH-PHEs and 1 + 2-/4-OH-PHE in Beijing was probably attributed to sources other than SHS.

Association between MDA and OH-PAHs. MDA was a product of lipid oxidative damage and hence was used as an indicator of lipid peroxidation.[15] In this study, MDA was detected in all the urine samples, and their median concentrations were 48.4 and 51.9 μg g^{-1} creatinine in Beijing and Los Angeles, respectively. No significant difference in the concentration of MDA was observed between the two cities (Figure 1 and Table 1). The relationship between MDA and OH-PAHs is shown in Figure S4. MDA was significantly correlated with Σ_{12}OH-PAHs ($p < 0.05$); however, for speciation analysis, only ΣOH-BPs was significantly correlated with MDA ($p < 0.05$). This result is out of our expectation because most species measured in this study were found to strongly associate with MDA or other oxidative damage

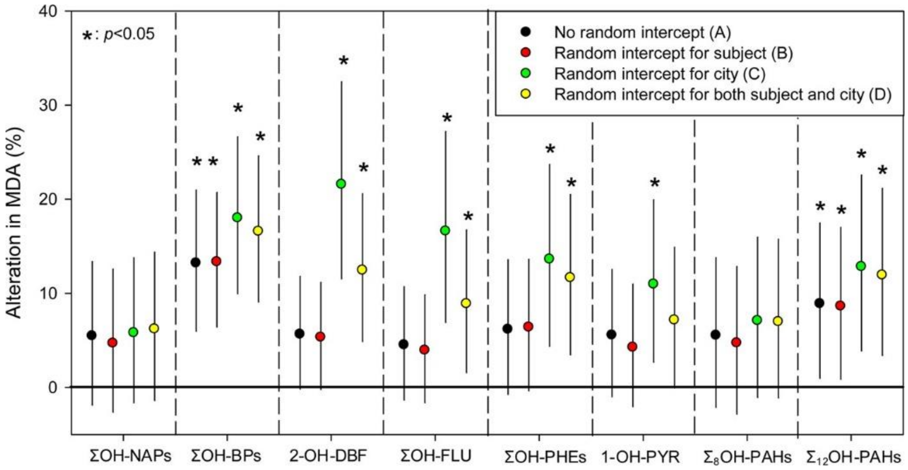


Figure 3. Association between OH-PAHs and MDA. The alteration in MDA (%) was associated with a one-fold increase of OH-PAHs.

Σ_8OH- PAHs: sum of ΣOH-NAPs, ΣOH-FLUs, ΣOH-PHEs, and 1-OH-PYR; Σ_{12}OH-PAHs: sum of Σ_8OH-PAHs, ΣOH-BPs, and 2-OH-DBF.

biomarkers (i.e., 8-hydroxy-2-deoxyguanosine and 8-isoprosta-glandin-F2α) as shown in previous studies.[6,14,46,47] In addition, the association between MDA and several OH-PAHs is marginally significant (Figure S4), suggesting there are some interference factors affecting the association.

To investigate the possible interference factors, we applied a simple linear regression model (Model A) and three mixed-effects models (Models B, C, and D) to study the association between MDA and OH-PAHs and then compared the results of different models. In Model A, the association between OH-PAHs and MDA was considered constant among individuals and cities, which is corresponding to the results in Figure S4. Among six OH-PAHs homologues, only ∑OH-BPs is significantly associated with MDA ($p < 0.05$). In Model B, the intercept was allowed to vary among subjects. As shown in Figure 3, the association between OH-PAHs and MDA was comparable with that in Model A, indicating that individual difference did not cause a significant interference in this study. In Model C, the intercept was allowed to vary between the two cities, and the results revealed a significant association between MDA and all OH-PAHs except for ∑OH-NAPs. Compared with Model A, the association between OH-PAHs and MDA was generally more significant, indicating that city is a major interference factor. The results of Model D, in which the intercept was varied among both subjects and cities, were similar to that of Model C, once again indicating a limited impact of individual difference compared with city.

As discussed above, the association between OH-PAHs and MDA was found to vary between the two cities, even for the same subject. There are several possible explanations for the observed city effect on associations: (1) the exposure to PAHs could induce the change in antioxidants in the human body,[48] which could affect an individual's oxidative stress; (2) the urinary MDA concentration was affected by other factors differing in two cities, such as the diet intake of MDA precursors and the decomposition conditions of MDA;[49] and (3) the cities' differences in the concentration of other pollutants that could induce oxidative damage[15] may interfere with the association between MDA and OH-PAHs. However, the potential mechanism of the observed city-effect is beyond the scope of this study and calls for future studies. Nevertheless, it is important to address that the associations between MDA and OH-PAHs are generally significant only if the city effect was considered. This is probably because OH-PAHs were significantly higher in Beijing but MDA was comparable between the two cities, which could weaken the inter-city associations (Figure S5).

There are several limitations of this study. First, the external exposures to PAHs were not measured, and thus, the OH-PAHs results could not be attributed to specific sources. For example, the time spent in indoor environments was not assessed in this study but may be an important factor affecting PAHs exposure levels and the related health effects.[50,51] Second, many factors (e.g., diet, stress, and physical activities, etc.) may have changed when the subjects traveled from Los Angeles to Beijing. How these factors affect OH-PAHs measured in this study is not fully understood. Finally, because the CYP enzyme cannot be readily measured in human subjects, how it affects the observed difference of the ratios of some OH-PAH isomers between the two cities cannot be determined.

In summary, this study identified significantly higher PAHs exposure and homogeneous OH-PAHs ratios in Beijing compared with Los Angeles in summer 2012. It also found a significant association between PAHs exposure and lipid peroxidation, with the association varying between the two cities. This study highlighted a possible link between PAH exposure and metabolism that needs to be considered in future health-effect studies.

ASSOCIATED CONTENT

Supporting Information

The Supporting Information is available free of charge on the ACS Publications website at DOI: 10.1021/acs.est.5b04629.

Tables showing information of OH-PAHs standards, the monitored ions of methylated OH-PAHs in GC-MS (EI) and the method detection limits of OH-PAHs, the influence of individual characteristic and physical activities on urinary OH-PAHs based on multivariate regression model with stepwise approach, comparison between the physical activities in Los Angeles and Beijing, descriptive statistics of biomarkers urine samples in Beijing (BJ) and Los Angeles before (LA1) and after the trip (LA2), Pearson correlation among different OH-PAHs and metabolite ratios, and summary of the studies cited to investigate the relationship between naphthalene exposure and 1-/2-OH-NAP ratio. Figures showing the concentration of grouped urinary OH-PAHs; concentration of urinary 1-OH-PYR in the population around world; a comparison in 1+2/4-PHEs, OH-PHEs, and time in secondhand smoke (SHS) among population in Los Angeles, in Beijing without SHS exposure, and in Beijing with SHS; correlation between MDA and OH-PAHs in both cities; and correlation between MDA and 1-OH-PYR in Los Angeles and Beijing. (PDF)

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Notes

The authors declare no competing financial interest.

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