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Comparing on-road real-time simultaneous in-cabin and outdoor particulate and gaseous concentrations for a range of ventilation scenarios

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Abstract

Advanced automobile technology, developed infrastructure, and changing economic markets have resulted in increasing commute times. Traffic is a major source of harmful pollutants and consequently daily peak exposures tend to occur near roadways or while traveling on them. The objective of this study was to measure simultaneous real-time particulate matter (particle numbers, lung-deposited surface area, $PM_{2.5}$, particle number size distributions) and CO concentrations outside and in-cabin of an on-road car during regular commutes to and from work. Data was collected for different ventilation parameters (windows open or closed, fan on, AC on), whilst traveling along different road-types with varying traffic densities. Multiple predictor variables were examined using linear mixed-effects models. Ambient pollutants (NO_x, $PM_{2.5}$, CO) and meteorological variables (wind speed, temperature, relative humidity, dew point) explained 5–44% of outdoor pollutant variability, while the time spent travelling behind a bus was statistically significant for $PM_{2.5}$, lung-deposited SA, and CO (adj-R^2 values = 0.12, 0.10, 0.13). The geometric mean diameter (GMD) for outdoor aerosol was 34 nm. Larger cabin GMDs were observed when windows were closed compared to open (b = 4.3, p-value = <0.01). When windows were open, cabin total aerosol concentrations tracked those outdoors. With windows closed, the pollutants took longer to enter the vehicle cabin, but also longer to exit it. Concentrations of pollutants in cabin were influenced by outdoor concentrations, ambient temperature, and the window/ventilation parameters. As expected, particle number concentrations were impacted the most by changes to window position / ventilation, and $PM_{2.5}$ the least. Car drivers can expect their highest exposures when driving with windows open or the fan on, and their lowest exposures during windows closed or the AC on. Final linear mixed-effects models could explain between 88–97% of cabin pollutant concentration variability. An individual may control their commuting exposure by applying dynamic behavior modification to adapt to changing pollutant scenarios.

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Supporting Information

This document provides information on the predictor variables used in regression analysis (Table S1), the basic descriptive statistics for measured outdoor and cabin pollutant concentrations (Table S2 and S3), the results for Pearson's product-moment correlation (r) (Table S4) and Spearman's Rank correlation (r_s) (for non-parametric data) (Table S5). It also presents photos of the experimental setup (Figure S1), and discusses the issue of self-pollution (Section 4.)

Keywords

Car commuting; exposure assessment; ventilation; pollution; aerosols

1. Introduction

Ambient air pollution is a complex mixture of primary and secondary organic and inorganic particulates and gases generated from both anthropogenic (combustion and non-combustion) and natural sources. Recent estimates have put the global annual mortality rate from ambient particulate matter (PM) pollution at more than 3 million, especially from cardiovascular and circulatory diseases, lower respiratory infections, chronic obstructive pulmonary disorder (COPD), and lung cancer, making it the 9th most important risk factor to human health globally, and the number one environmental risk factor (Lim *et al.*, 2012). The positive association with lung cancer lead, in 2013, to the International Agency for Research on Cancer (IARC) Working Group to unanimously classify outdoor air pollution as carcinogenic to humans (Group 1); PM was evaluated separately and also classified as Group 1 (Loomis *et al.*, 2013). In developed countries, transport contributes 25–40% to ambient pollutant concentrations collectively, although for some pollutants such as carbon monoxide (CO), nitrogen oxides (NO_x), and ultrafine particles (UFP), traffic may contribute up to 90% (Keuken *et al.*, 2005; Greenbaum, 2013). In fact, diesel and gasoline exhaust have both been classed by the IARC as Group 1 and Group 2B carcinogens, respectively(Russell, 2013). Because traffic emissions are a major source of pollutants, peak concentrations tend to occur near or on roads, which is where an individual may receive a disproportionately large fraction of their total daily personal exposures. Indeed studies have reported elevated risks for developing asthma and reduced lung function in children living near to heavily-trafficked roads (Brugge *et al.*, 2007), as well as measured changes in cardiac biomarkers and pulmonary function in adults driving or working in private vehicles (cars) (Riediker *et al.*, 2004; Heinrich *et al.*, 2005; Sarnat *et al.*, 2014).

Cars continue to dominate the commuting landscape in the US. According to the 2009 National Household Travel Survey (NHTS) conducted by the US Department of Transport (USDOT) (Usdot, 2009), 91% of US commuters travel an average of 24.4 miles (46 minutes) to and from work by private vehicle; the vast majority (~80%), commute alone (Mckenzie, 2015). Indeed, private vehicle ownership has continued to rise since surveys began in 1969, so that by 2009 the number of personal vehicles far exceeded the number of drivers. In addition, the average age of the US vehicle fleet has also increased so that 40% of all private cars are now 10 years of age or older (Usdot, 2009). This has potential environmental and health consequences given that older vehicles tend to generate higher emissions, due to age-related deterioration of vehicle control systems (Borken-Kleefeld e Chen, 2015), impurity-enriched lubricating oil in the crankcase (Russell, 2013), abrasion and wear and tear of metallic components (Greenbaum, 2013), and generally more permissive emission standards (Krasenbrink *et al.*, 2005). The persistent lack of investment and public support continues to hamper the development of transport alternatives, thus vehicle ownership and traffic flows are only expected to increase.

Many laboratory and field studies have examined pollutant concentrations and commuter exposures during car travel (Kaur *et al.*, 2007; Zuurbier *et al.*, 2010; De Nazelle *et al.*, 2012; Kingham *et al.*, 2013; Ragettli *et al.*, 2013; Suarez *et al.*, 2014; Good *et al.*, 2016). Factors such as vehicle age, fuel type, driving patterns (acceleration / idling), vehicle speed, proximity to other cars, road-type, position on the road, self-pollution, traffic mix, meteorology, topography, and road condition, have all been reported to influence the local pollutant environment outside of a vehicle (Van Wijnen *et al.*, 1995; Knibbs *et al.*, 2010; Knibbs *et al.*, 2011; Kingham *et al.*, 2013). And although studies investigating car commuting exposures have highlighted elevated cabin pollutant concentrations whilst traveling through tunnels and on freeways (Kaminsky *et al.*, 2009; Knibbs *et al.*, 2010), with both increasing and decreasing vehicle speeds (Hudda *et al.*, 2012; Ding *et al.*, 2016), and higher road and traffic densities (Weichenthal *et al.*, 2015), the air exchange rate (AER) (dependent on ventilation parameters such as window position, the ventilation system (fan/ AC), natural leakage from door seals and window cracks) is highlighted as among the most important determinants of cabin concentrations, or cabin particle removal (Hudda *et al.*, 2011; Knibbs *et al.*, 2011). Kaminsky et al., (2009) observed the highest UFP concentrations with AC followed by windows open then windows closed, while Hudda et al., (2012) and Ding et al., (2016) reported lower indoor / outdoor ratios when the fan was operating under re-circulation (compared to non-recirculation) mode. The duration of the commute has also been highlighted when cumulative exposures were considered (Good *et al.*, 2016), something that is often neglected when averages are the main metric with which results are described.

Only a few studies have previously obtained simultaneous in-cabin / outdoor measurements. Hudda and Fruin (2013) collected multiple particle metrics (UFP, $PM_{2.5}$, PM_{10}, black carbon (BC), particle-bound PAHs) during 3 ventilation modes: fan off, fan on (recirculation mode), fan on (outside air). They also conducted earlier studies measuring particle number size distributions (PNSD) and total particle number concentrations (Hudda *et al.*, 2011; Hudda *et al.*, 2012). Zhu et al., (2007) also measured in-cabin / outdoor PNSD and particle number concentrations in 3 vehicles for the same ventilation modes as the previous study. Bigazzi and Figliozzi (2012) measured UFP in 3 vehicles for a range of ventilation scenarios, however simultaneous in-cabin / outdoor measurements were only collected during the latter part of the study. Finally, Weichenthal et al., (2015) measured simultaneous in-cabin / outdoor UFP, BC, CO, and $PM_{2.5}$ concentrations in 3 cities in Canada in one vehicle type; however, the different ventilation scenarios were not investigated.

Existing studies provide invaluable insights into the factors controlling cabin concentrations. However, one of the main objectives and largest contributions of this work was to collect / present simultaneously collected in-cabin and outdoor data for multiple pollutant metrics during realistic car commutes. To the authors' knowledge, no other study has incorporated real home-to-work-to-home commutes, thus capturing a real-world, non-contrived commuting experience, as well as related complexities. The next objective was to examine the parameters effecting both outdoor concentrations and outdoor and cabin associations. This study is also unique in conducting regression analysis on potential variables influencing the air directly outside of a vehicle, before then trying to ascertain how these variables influence the air inside of the vehicle. Four commonly used ventilation parameters were

investigated: windows open, windows closed, fan on, AC on, and rigorous analysis was performed using linear regression and mixed-effects models. No other study has previously measured these 4 ventilation scenarios using multiple pollutant metrics, and results will show how they impact pollutants differently. This is also the first commuting study to incorporate lung-deposited surface area (SA) measurements, despite being increasingly cited as the particle metric which correlates the most strongly with inflammatory responses (Oberdorster, 1996; Brown *et al.*, 2001; Karakoçak *et al.*, 2016). It is also the first study of its kind in the Midwest, in spite of the overwhelming reliance of the private motor vehicle in this region. The overall aim of this study was to identify when and during which ventilation scenarios commuters may receive their highest exposures, and how this may be mitigated through behavior modification.

2. Methods

2.1 Site / location

Measurements were collected along 2 routes from Washington University in St Louis' south-east parking lot (work destination) to 2 residential locations: one in Shaw (63110), an inner-city residential neighborhood with a grid layout approximately 6 miles south-east of the university, and one in Hazelwood (63042), a residential / industrial suburban location, around 18 miles north-west of the university and 4 miles north of Lambert International airport (Figure 1). These routes comprise both freeways and non-freeway roads with varying aspect (width-to-height) ratios and surroundings. They also pass several industries including an aircraft manufacturing facility, a mail shipping center, a pharmaceutical and chemical business, a coffee roasting factory, and various stores, restaurants, and fast-food eateries. Traffic densities differed depending on the road-types being traversed and the time of day.

2.2 Experimental plan

Real-time simultaneous particulate matter and CO measurements were collected outside and inside of an on-road car operating under different ventilation scenarios: windows open, windows closed, fan on (midway, no recirculation), AC on (midway, no recirculation). When either the fan or AC were operating, the windows were closed. Two vehicles with gasoline-fueled internal combustion engines and cabin filters were used in this study: a Hyundai Sonata 2013 (Car 1), and a Subaru Outback 2010 (Car 2). Car 1 had driven 18,500 miles since the cabin filter had been changed, and the Hyundai, 15,000 miles. Each vehicle was fitted with five pairs of instruments. The first set of instruments sampled outdoor air: the inlet was located 3 cm outside of the back left passenger window. A 2 cm strip of foam was fitted into a crack in the window with feed-through for the tubing and a CO sensor. A second set of identical instruments sampled cabin air: this inlet was located in what would be considered the breathing zone of a front seat passenger. A schematic of the experimental setup is presented in Figure 2, while photographs of the cars and experimental setup are provided in the Supporting Information (Figures S1). Minimal amounts of conductive silicone tubing were used for all aerosol instruments to minimize particle losses, and all tubing lengths were replicated for each set of instruments so that results would be comparable. No tubing was necessary for the CO sensors. The instrument setup was identical within each vehicle.

Data from a total of 56 commutes were collected across 25 non-consecutive days from September – November, 2014, between 8–10am, and 4–7 pm - capturing at least some of the morning and evening rush hours. Due to instrument and personnel limitations, sometimes more than 2 commutes were performed in one day. Each ventilation scenario was measured a minimum of 12 times. A dashboard surveillance camera (dashcam) (Figure 2) was also installed on the front windshield to provide real-time geopositioning information (location and speed). This enabled traffic density and type to be recorded and categorized post hoc, and pollutant data to be matched with precise vehicle location and surrounding traffic. Other relevant events were also either captured visually – such as times the vehicle was following a bus or truck, or communicated verbally via the driver, such as vehicles travelling to the left of the measurement vehicle (close to the outdoor inlet), or smells - cigarette smoke, or food from restaurants. Meteorological data (temperature, wind speed, relative humidity, dew point and wind direction), and ambient pollutant data (NO_x, $PM_{2.5}$, CO) are known to influence urban particulate concentrations (Hitchins *et al.*, 2000; Charron e Harrison, 2003; Janhäll *et al.*, 2004; Zhang e Wexler, 2004; Kuhn *et al.*, 2005; Longley *et al.*, 2005; Westerdahl *et al.*, 2005; Hussein *et al.*, 2006; Leavey, 2009) and were collected post-hoc from nearby fixed site monitors (FSM), operated by NOAA's National Climate Data Center (NCDC) and the US Environmental Protection Agency (USEPA), respectively (Ncdc, 2015; Usepa, 2015) to be used in subsequent regression analysis. A complete list of the explanatory variables collected in this study is provided in the Supporting Information (Table S1). Given that one inlet was outside without shelter, no measurements were collected when it was raining.

2.3 Instruments

All instruments were light-weight, portable, and battery-powered with fast response times so that changes in pollutant concentrations could be easily captured. A P-TRAK Ultrafine Particle Counter 8525 (TSI, Inc.) measured particle numbers. It uses isopropyl alcohol to grow the particle, and condensation particle counter (CPC) technology to count particles in the size range 0.02 to 1 μm for concentrations up to 5×10^5 #/cc. A DustTrak II Aerosol Monitor 8532 (TSI Inc.) measured particle mass ($PM_{2.5}$) concentrations up to 150 mg/m^3 using the principles of light-scattering. Lung-deposited surface area (SA) was measured using an AeroTrak 9000 Nanoparticle Aerosol Monitor (TSI Inc.), reporting the fraction of SA of particles in the size range 0.01 to 1 μm^2/cm^3 that would deposit in different regions of the respiratory tract according to the IARC. A more complete description is provided in Leavey et al., (2013). For this study, the fraction of particles depositing in the alveolar (A) region was collected. A Portable Aerosol Mobility Spectrometer (PAMS) (Kanomax, Japan, Inc.) was used to measure particle number size distributions (PNSD) in the size range 0.014 to 0.863 μm. PAMS uses a bipolar charger to charge particles which can then be classified using a differential mobility analyzer (DMA) (Qi e Kulkarni, 2016), and a CPC to count these classified particles (Kulkarni *et al.*, 2016). Finally, a Lascar EL-USB-CO300 sensor (Lascar Electronics) was used to measure CO with one reading every 10 seconds for concentrations between 0–300 ppm. All instrument and laptop timestamps were synchronized to the dashcam time prior to each measurement, to ensure that location and pollutant data were matched. Zero calibration and maintenance protocols were followed between samples. All instruments had been recently calibrated by the manufacturer. In addition, they were collocated and calibrated in the laboratory before and after the

measurement study, using sodium chloride generated from a nebulizer. Only minimal correction factors (<5%) were applied to the older Ptrak, AeroTrak, and DustTrak to facilitate comparisons.

2.4 Data analysis

All instrument data were imported into TrakPro software (version 4.6.1.0, TSI Inc.) and subsequently exported into Excel 2013, where they were checked manually and cleaned. Although a total of 56 commutes were made, one was abandoned due to rain. Another 4 commutes only collected particle number concentrations (P-TRAK) and particle number size distributions (PAMS). Instrument malfunctions caused some data losses. The final dataset was comprised of between 47–51 samples for every instrument. A matrix was generated and imported into R version 3.2.5 (2016-04-14) Copyright © 2016 The R Foundation for Statistical Computing), for statistical analysis. The following R packages were also used: lme4 (Bates *et al.*, 2015), car (Fox e Weisberg, 2011), ggplot2 (Wickham, 2009), nortest (Gross e Ligges, 2015), matrix (Bates e Maechler, 2016), sjmisc (Lüdecke, 2016a), and sjPlot (Lüdecke, 2016b). All graphics were generated in SigmaPlot 11.0.0.75 Copyright © 2008 Systat Software, Inc.

Many statistical tests require data be normally distributed. All continuous variables were tested for normality using a range of graphical analysis tools (histograms, boxplots, stem plots, qq-plots), combined with formal hypothesis tests (Shapiro-Wilk and Anderson-Darling). Those that did not satisfy these tests of normality were log-transformed and retested. All dependent variables were positively skewed and were log-transformed, thus ensuring normality. Analyses were conducted on data for the whole car journey. Because the CO data contained many zero data points they were problematic to transform, and so the total area under the curve was selected as the dependent variables' statistical metric to examine both cabin and outdoor concentrations for all pollutants, log-transformed to ensure normality (Osborne e Waters, 2002), with the exception of PNSD where GMD was used.

Regression analysis was the primary method of statistical testing used in this study to ascertain the factors influencing measured outdoor and cabin pollutant concentrations. However, samples collected on either the same or consecutive days may have more in common with each other than with other days. This may have an "unpredictable" and "random" influence on the data (Winter, 2013). Therefore linear mixed-effects models (LME), which are extensions of linear regression models and fit by maximum likelihood, were built for each dependent pollutant variable, with instruction from several tutorials (Winter, 2013; Iqss, 2016; Lüdecke, 2016b). The fixed-effect was one independent variable, and the random-effect (intercept) was day (i.e. the day of data collection). For multiple predictors, each model started with the random intercept variable day, while the fixed-effects were left empty (null model). Next, one new explanatory fixed-effects variable was added to the model (newer model), and the output was then compared to the previous (reduced) model using the Likelihood Ratio Test approach (Winter, 2013). If the likelihood of the newer model was significantly different from the reduced model ($p=<0.05$), then the effects of this additional explanatory variable were significant and retained as a fixed-effect. This methodology was repeated until the model could no longer be improved. All models for the

outdoor pollutants included the journey duration variable, which was used to control for the different commute times.

3 Results and Discussion

3.1 Descriptive statistics

The final dataset was comprised of 55 commutes: 20 for Car 1 and 35 for Car 2. Twenty-one of these commutes were in the morning, and the remaining 34 were in the afternoon. Average temperature over the course of the field campaign was 66.9 °F (SD 12.0), while wind speeds ranged from 1.5 to 20.7 m/s; no particular wind direction dominated. Background nitrogen oxide (NO_x) concentrations, a good indicator of general air quality in urban areas, also fluctuated between 2.5–58 ppb. Descriptive statistics were calculated for all pollutant metrics for the entire journey, and results are presented in Tables S2 and S3 of the Supporting Information. In addition, a matrix of both Pearson's product-moment correlation (r) and Spearman's Rank correlation (r_s) (for non-parametric data) is also presented (Tables S4 and S5). However, it is worth highlighting here that the average geometric mean diameter (GMD) of measured outdoor PNSD was 34 nm (minimum 23 nm, maximum 52 nm). Similar, though frequently lower GMDs, are reported in the literature, and tend to be affected by road incline/decline, vehicle speeds, meteorology, distance from source, and sulfur content of the fuel (Palmgren *et al.*, 2003; Kittelson *et al.*, 2004; Zhang e Wexler, 2004; Dahl *et al.*, 2006; Cheng *et al.*, 2010; Yamada *et al.*, 2016). The relatively high lower cut-off of the instrument (14 nm) might explain the higher GMD reported here. The following sections will seek to identify the most important parameters effecting a) outdoor and b) cabin pollutant concentrations during a commute. The influence of window position / ventilation scenarios (windows open, windows closed, fan on, AC on) on the outdoor / cabin correlations will also be examined using time-series, and regression analyses with linear mixed-effects models.

3.2 Outdoor pollutant concentrations

The pollutant concentrations present directly outside of a vehicle during a commute are a combination of ambient pollutant concentrations and pollutant concentrations from nearby sources such as traffic and self-pollution, both of which are modified by meteorological, geological and topographical conditions. The fixed-effects terms for each LME model was one of the selected independent variables (i.e. NO_x, wind speed etc. Table S1), controlled for journey duration. The random-effects term was the measurement day.

Table 1 presents the results of simple regression analysis for all variables demonstrating a significant association with measured outdoor pollutant concentrations (log area under the curve) at the 90^{th}, 95^{th}, or 99^{th} % confidence interval (CI). For PNSD, the GMD was the dependent variable. In general, the pollutant data collected from a nearby fixed-site monitor, and representing ambient pollutant conditions log(NO_x, $PM_{2.5}$ and CO) demonstrated the highest positive correlations with outdoor commuting concentrations. Ambient log(NO_x) proved the strongest predictor of particle number concentrations (#/cc) with an adj-R^2 of 0.36, which is not surprising given they share a common principal source in urban areas: vehicular emissions (Aqeg, 2005). Perhaps again unsurprisingly, ambient log($PM_{2.5}$) was the

best ambient pollutant metric to represent commuting $PM_{2.5}$ (μg/m^3), explaining about 33% of the variance. Lung-deposited SA (μm^2/m^3) was explained to some degree by all 3 ambient metrics (with adj-R^2 values of between 0.24 – 0.31), again highlighting traffic as a major source; while CO (ppm) correlated only with ambient log($PM_{2.5}$). While a major source of CO and NO_x in urban areas are engine emissions from automobiles, $PM_{2.5}$ is derived from both engine and non-engine sources if measured close to traffic (Palmgren *et al.*, 2003; Thorpe e Harrison, 2008), but also from crustal matter, dust, and long-range transport as well (Querol *et al.*, 2001; Hueglin *et al.*, 2005). No correlation between the ambient pollutants and GMD was observed. These findings highlight that ambient pollutants may not represent local pollutant metrics equally, thus they should be selected carefully depending on which pollutant metric is of most concern.

The majority of meteorological variables also proved to be significantly correlated with outdoor commuting particulate pollutants, for example increasing wind speed was associated with a decrease in particle numbers, lung-deposited SA, and to a certain extent particle size (GMD) (adj-R^2 = 0.25, 0.28, and 0.05 respectively). This is likely due to increased dilution and mixing with the cleaner background air (Shi *et al.*, 1999; Hitchins *et al.*, 2000; Zhu and Hinds, 2005), as well as increased transport and deposition rates (Zhang and Wexler, 2004). According to Pirjola et al (2006), the correlation between particles and wind speed is especially strong within 10 m of a roadside, primarily for particles below 100 nm, while Charron & Harrison (2003) only reported correlations for particles 30–450 nm in size. Ambient temperature demonstrated a negative association with particle numbers – with higher particle concentrations at lower temperatures (adj-R^2 0.18). Temperature also correlated, this time positively, with PNSD, so that for every 1 unit increase in temperature there was a 0.31 nm increase in GMD (adj-R^2 of 0.18; p-value 0.02). These trends may be explained in part by: 1) increased coagulation and diffusion in warmer temperatures in part from the increased vertical mixing of air masses (Shi *et al.*, 1999; Hussein *et al.*, 2006), causing a shift to larger particles and thus fewer numbers; 2) decreased secondary particle formation from the condensation of precursor gases (Wehner *et al.*, 2002), and 3) a reduction in the rate of condensation growth of fresh particles (Yao *et al.*, 2007). A positive correlation was also observed for $PM_{2.5}$, which has been associated in previous studies with increased sulfate formation in warmer temperatures from SO_2 oxidation (Ito *et al.*, 2007; Tai *et al.*, 2010), and atmospheric stagnation (Tai *et al.*, 2010), and in a more localized setting decreased brake-pad efficiency and increased dust generation (Grigoratos e Martini, 2014). Different wind direction patterns in warmer temperatures have also been reported (Ito *et al.*, 2007); indeed in this study easterlies dominated during warmer temperatures, and higher $PM_{2.5}$ concentrations were measured during these easterlies (Table 1). It is important to add that driving behavior, traffic-type, and surface-to-tire interactions may also differ during warmer temperatures.

The time spent (seconds) travelling behind a bus (TimeBB, Table 1) was positively associated with $PM_{2.5}$, lung-deposited SA, and CO. This is not surprising as old diesel buses are considered gross-polluters, and many studies have linked them with elevated on-road pollutant concentrations (Dennekamp *et al.*, 2002; Weijers *et al.*, 2004; Kaur *et al.*, 2007; Briggs *et al.*, 2008). However, the associations between the explanatory traffic variables - traffic density (0,1,2), presence of buses / trucks (0,1), direction (0,1) and the measured

outdoor pollutants were generally weak, and may come down to classification issues, for example: 1) It was difficult to reduce an entire commute into a single traffic-density class, because the commutes were comprised of many roads with different traffic densities. 2) Commuting traffic was often unidirectional and many of the commutes were counter-traffic congestion. For instance, the drive home from work was often moderate traffic but the freeway on the other side of the road was completely jammed. Therefore allocating a moderate classification to the commute may neglect the potential influence of counter traffic. 3) Wind variables, especially wind direction, may influence the degree that the other side of the road is influencing the road commuted. 4) Siegmann et al., (2008) discusses "superpolluters" – attributing much of the pollution emitted locally to a small percentage of poorly maintained vehicles. Therefore, a relevant traffic variable may focus less on traffic density, and more on the *type* of surrounding traffic, traffic mix, or its upkeep. As such, a car commuter's exposure may be influenced more by the type of traffic in the immediate surroundings than the density of traffic and route selection. Of course, the chance of interacting with a superpolluter is increased with a higher traffic density, and no examination of the effects of idling and acceleration with increasing traffic density was conducted. The traffic density variable was also compared to the ambient pollution variables log(NO_x, $PM_{2.5}$, and CO), which had correlated fairly well with measured outdoor pollutant concentrations. While ambient log(NO_x) and log(CO) demonstrated some correlation with traffic density (R^2 = 0.04 and 0.06, p-values = <0.07), when regressions were controlled for the time of day, then traffic was no longer important, thus reinforcing the points made previously. Although pollutant concentrations were generally higher during morning commutes compared to afternoon, once the regressions included the ambient pollutants, this variable was no longer significant. Finally, no statistical difference between the routes driven by the 2 cars were observed. When the linear mixed-effects models were generated using one ambient pollutant metric and one meteorological metric, the random variable day was able to explain 32%, 45%, 45%, 6%, and 19% of the random variability for particle numbers, lung-deposited SA, $PM_{2.5}$, CO, and GMD, respectively (Iqss, 2016). In general, $PM_{2.5}$ was the easiest pollutant metric to predict, and CO and GMD the most difficult.

3.3 In-cabin and outdoor time-series

The influence of window position and ventilation parameters on cabin / outdoor correlations was a key focus of this study. The following section compares two scenarios: a) windows open, and b) windows closed, for the different pollutant metrics, highlighting where the similarities and differences lie. The influence of window position as well as the fan and AC will also be examined during subsequent regression analysis.

Figures 3(a–h) presents multiple time-series of the 4 pollutant metrics depicting simultaneous outdoor and cabin concentrations for windows open and closed. All commutes began in a fairly quiet environment (university or residential parking lot / private drive) where lower concentrations are expected, before progressing through varying concentration fields (including freeways, street canyons, multiple traffic lights, and cross roads), before terminating again in a quiet environment. Outdoor concentrations are a combination of both local sources and ambient conditions (traffic, weather, topography, other sources), while cabin concentrations are typically the product of outdoor concentrations which have been

subsequently modified by the presence of the vehicle cabin and AER. Contamination due to self-pollution was minimal (see Supporting Information Section 4 for more information). In general, when windows were open cabin concentrations tracked outdoor concentrations, albeit with some reduced variability and slight time lags. This is especially true for particle number counts, and to a lesser extent CO. Particle numbers also demonstrated the highest outdoor variability of the particle metrics, highlighting the high spatiotemporal variability of UFP (<100 nm), the dominant size of particle numbers close to source (traffic). At times cabin concentrations even exceeded outdoor concentrations, while other times outdoor spikes failed to influence cabin concentrations at all, which is likely due to the short duration of the outside elevation.

In contrast, closed windows generally produced a smoothing effect on cabin concentrations, with pollutants demonstrating reduced variability, fewer in-cabin spikes, and more pronounced time-lags between outdoor-to-cabin elevations, so that as the vehicle passed through a high concentration environment the cabin concentrations slowly rose, but equally as the vehicle returned to a lower concentration environment the cabin concentrations remained elevated, taking a longer time to dilute/diffuse, see Figure 3g (CO). It should also be noted that cabin $PM_{2.5}$ and lung-deposited SA profiles frequently differed from those of particle counts and CO, often exhibiting elevated cabin concentrations, at least for periods of time, which was exacerbated when windows were closed. This will be examined in more detail in further work. Figure 4(a–b) presents cabin and outdoor PNSD averaged across a single commute. Distributions were bimodal with outdoor mode diameters around 10–30 and 90–110 nm (compared to a mean GMD for all runs around 34 nm), with slightly larger cabin GMDs when windows were closed. According to Zhu et al (2007), smaller GMDs occur when sampling roads with higher diesel emissions which in turn are modified to a greater extent by ambient temperature and relative humidity. However, no information on diesel versus gasoline emission ratios were collected in this study. Finally, Figure 4(c–j) displays 20, 60, 100, and 250nm particle counts, for windows opens and closed. Similar to previous observations, cabin particle size and concentration track outdoors most closely when windows are open, while larger cabin sizes and reduced variability are observed when windows are closed. The degree to which cabin concentrations are influenced by outdoor concentrations depends on the AER and the particle penetration factor which is dependent on particle size, with higher ratios generally reported for larger particles (Zhu *et al.*, 2005); however more data is required before conclusions for this study can be drawn.

3.4 In-cabin pollutant concentrations

This section provides a more rigorous assessment of commuter exposure by examining cabin pollutant concentrations using LME. The dependent variables were the log-transformed measured cabin pollutant concentrations, the fixed-effect variables were the log-transformed outdoor measured concentrations, and the categorical ventilation variable: windows open (reference level), windows closed, fan on, AC on. The random intercept variable was day. Models were not controlled for journey duration because the outdoor pollutant variable was now included. Results demonstrated that cabin pollutants were predominantly influenced by the outdoor concentrations, which in this study were able to explain 64%, 28%, 92%, 28%, and 16% of cabin variability for particle numbers, lung-deposited SA, $PM_{2.5}$, CO

concentrations, and GMD, respectively (p-values <0.01). Lung-deposited SA demonstrated lower cabin concentrations compared to outdoor (b = 0.5257, p-value <0.01) while the size of cabin particles tended to be slightly smaller than outside (b = 0.71, p-value = <0.01).

However, the vehicle's ventilation scenario exerted a significant influence on cabin / outdoor correlations for all pollutants (Figure 6). For particle numbers, cabin concentrations tracked outdoor concentrations most closely when windows were open (reference level). Associations were significantly modified when the fan was running (b = −0.217), and even more so when the AC was running (b = −0.416), or the windows were closed (b = −0.563) (p-values <0.05). This ventilation variable explained 7% of the total cabin variation in particle numbers. In fact, running the AC or keeping the windows closed provided such a protective influence that cabin / outdoor correlations were no longer significantly correlated. A similar trend was also observed for lung-deposited SA. While the ambient variable temperature was not significant for outdoor lung-deposited SA concentrations, it was a strong predictor of cabin SA concentrations with higher concentrations measured in warmer temperatures (b = 0.04, p-value = <0.01). This may indicate some secondary organic aerosol (SOA) formation due to the correlation between ambient temperature and ambient ozone, and ambient ozone and SOA (Lamorena and Lee, 2008), although this was not observed for any other measured pollutant. Cabin $PM_{2.5}$ concentrations tracked measured outdoor concentrations the most closely, and were the least influenced by any change in ventilation parameters (<1%). Cabin CO concentrations were lower than their outdoor counterparts (b = 0.889, p-value <0.01), and modified the most when windows were closed, closely followed by running the AC. In fact, for 33% of journeys, no cabin CO was recorded, and 75% of these cases were observed when either the AC was running or windows were closed, again highlighting the protective effect of these ventilation parameters. Running the fan generated cabin concentrations that tracked outdoor concentrations the most closely (b = 0.875) but results were not statistically significant. Larger cabin particle sizes (GMD) were observed when windows were closed compared to open (b = 4.3, p-value <0.05), which may be explained by a lower penetration factor and higher deposition rates for particles <100 nm (Xu *et al.*, 2010), and reduced dilution permitting increased time for coagulation (Cyrys *et al.*, 2004). Finally, the effect of the car make was examined to establish whether the cabin / outdoor correlations and ventilation parameters were different between cars. None was observed for any of the pollutants. The final LME models presented in Figure 5a–d were able to predict 97%, 88%, 96% and 90% of the measured cabin concentrations for particle numbers, lung-deposited SA, $PM_{2.5}$, and GMD, respectively. The LME model for CO was unstable and was not included in the final figure. All final models satisfied the normality requirements during residual diagnostics (residual plots, histograms, qq-plots). Satisfactory validation of these models was also completed (results not shown).

When either the fan or AC were operating, air was not being recirculated in the vehicle cabin. However, the AC consistently resulted in lower cabin concentrations compared to the fan. The process of thermophoretic deposition is proposed as a possible explanation. When air enters the vehicle duct, it passes through the filter and then a fan which accelerates the air. Next, it passes through an evaporator which, when the AC is operating, cools the air depending on the temperature setting, the amount of Freon in the system, and ambient temperature and humidity, before entering the vehicle cabin via the vents. When the fan is

operating, the outdoor air follows the same trajectory with the only exception being that the evaporator is no longer cooling the air. The increased temperature gradient that exists between ambient air and the evaporator surface when the AC is operating results in a higher thermophoretic velocity and subsequently a higher particle deposition rate, i.e. more particles are depositing in the duct and so fewer particles are entering the cabin. The same explanation may be used to explain the higher GMD observed when the AC was operating where again the increased temperature gradient causes a higher thermophoretic velocity and particle deposition rate especially for the smaller particles.

There are many factors that complicate cabin / outdoor pollutant correlations. 1) The location of a source (in front of / adjacent to car) may impact the cabin and outdoor inlets differently, as will the lane the measurement vehicle is in (its traffic density, proximity to other lanes, proximity to moving traffic). 2) Vehicle speed will affect the generation of tail-pipe and non-engine emissions (resuspension from mechanical turbulence) (Kittelson *et al.*, 2004; Dahl *et al.*, 2006), the infiltration of particulates to the cabin by altering the AER of the vehicle (Xu *et al.*, 2010; Hudda *et al.*, 2011; Knibbs *et al.*, 2011; Ding *et al.*, 2016), and how long a vehicle spends in a given location (see point 4 below). Because speed is dynamic, it was too difficult to capture for the whole commute; however, its importance will be examined in future work. 3) Wind speed and direction will affect how emissions are diluted, and whether they are channeled towards or away from the measurement vehicle; it may also influence the penetration of these emissions into the vehicle. 4) Time-scale: the longer a vehicle is exposed to high outdoor concentrations the more they may penetrate the vehicle cavity. This can be observed in the time-series in Figure 3. If a vehicle passes only briefly through a polluted environment, its cabin may offer protection from the pollutants; conversely if it spends a prolonged time in elevated concentrations, the cabin may trap pollutants inside of the vehicle. This again is dependent on AER and ventilation parameters. 5) Self-pollution from the engine, resuspension of dust inside the vehicle, and isopropyl emissions from the 4 particle number instruments may have also complicated the cabin and outdoor correlations (See Supporting Information). Some contribution from SOA is also likely, especially given the higher cabin concentrations sometimes observed at the start of the commute (which is not believed to be related to instrument emissions).

While there are smart cars currently on the market with HEPA filters installed to remove particulates including pollen and viruses, most vehicles are not equipped with such efficient removal devices. This study demonstrates that a driver may still be able to control their commuting exposure by being cognizant of their pollutant environment and applying dynamic behavior modification to adapt to changing scenarios. For example, when a driver enters a high pollutant environment they could turn on the AC to reduce intake of the dirty air; when they enter a clean environment they could open their windows thus flushing pollutants from the vehicle. Of course, knowing when the air is clean or dirty is not always intuitive. Although much research is currently being conducted on mapping street pollution using multiple distributed wireless sensors, or disseminating local air quality along with local weather using a mobile app, a driver's ability to access information on the real-time air quality of a specific driving route is presently beyond the grasp of most commuters. What is more, this data will not provide information on what is actually occurring inside of a vehicle. Perhaps in the future cars may be equipped with pollutant sensors that can measure the

outdoor air and automatically apply mechanisms to prevent its infiltration indoors. Until that technology is available, further examination of the data collected during this campaign will be performed with the aim of examining the different roadway microenvironments, and providing some interim solutions to reduce commuter exposure.

4. Conclusion

The objective of this study was to collect simultaneous real-time particulate and CO measurements outside and inside of an on-road car, to identify the parameters effecting both outdoor concentrations and outdoor and cabin associations, and to examine when commuters receive their highest exposures and how this may be mitigated. Four ventilation parameters were investigated: windows open, windows closed, fan on, AC on, and rigorous analysis was performed using linear regression and mixed-effects models. The final dataset was comprised of 55 commutes. Ambient NO_x and $PM_{2.5}$ were important predictors of pollutant concentrations outside of a moving vehicle, explaining 18–36% of variability, while the meteorological variables (for example wind speed and temperature) explained between 5–44%. Of the traffic variables, only the time spent travelling behind a bus was significantly significant for $PM_{2.5}$, lung-deposited SA, and CO. The lack of significance for the other traffic variables is likely due to coding difficulties and this will be examined in further work. Time-series analysis demonstrated that cabin concentrations tended to track measured outdoor concentrations albeit with some reduced variability and time-lags when windows were open. When windows were closed, a smoothing effect occurred so that whether a pollutant entered the cabin was in part due to the outside pollutant concentration and the time spent in that elevated concentration. In general, pollutants took longer to enter the vehicle, but also longer to exit it. The vehicle cabin may therefore be viewed as acting both as a buffer - reducing the penetration of pollutants into the vehicle, and as a sink – capturing pollutants and slowing dispersion. In-cabin pollutants were predominantly influenced by the measured outdoor concentrations, which were able to explain 64%, 28%, 92%, 28%, and 16% of cabin variability for particle numbers, lung-deposited SA, $PM_{2.5}$, CO concentrations, and GMD, respectively (p-values <0.01). The vehicle's ventilation parameters, which effect the AER and thus particle penetration or removal, were also important. Cabin pollutant concentrations tracked outdoor concentrations most closely when windows were open or the fan was running. Commuting with the windows closed or the AC on reduced cabin concentrations significantly enough to disrupt cabin / outdoor correlations for the majority of measured pollutants, and frequently prevented the diffusion of CO into the vehicle. Running the AC also significantly increased the mean size of particle numbers measured inside the vehicle. Particle number concentrations were the most effected by changing ventilation scenarios, with $PM_{2.5}$ largely immune to such changes. In general however, car drivers can expect their highest exposures when driving with windows open or the fan on, and their lowest with windows closed or the AC running. A driver may be able to control their commuting exposures by being cognizant of their pollutant environment and applying dynamic behavior modification to adapt to changing scenarios. A more detailed examination of these changing pollutant scenarios in different roadway microenvironments will be conducted in follow-up work in an attempt to further identify important parameters influencing a commuter's exposure, from which mitigation strategies can be developed.

Supplementary Material

Refer to Web version on PubMed Central for supplementary material.

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References

AQEG, A. Q. E. G.. Particulate Matter in the United Kingdom. Department of Environment, Food and Rural Affairs (DEFRA); London: 2005. p. 1-444.

BATES, D., MAECHLER, M. [Acesso em: 10-04-16] Sparse and Dense Matrix Classes and Methods. 2016. Disponível em: < http://Matrix.R-forge.R-project.org/ >

BATES D, et al. Fitting Linear Mixed-Effects Models Using lme4. Journal of Statistical Software. 2015; 67(1):1–48.

BIGAZZI AY, FIGLIOZZI MA. Impacts of freeway traffic conditions on in-vehicle exposure to ultrafine particulate matter. Atmospheric Environment. 2012; 60:495–503.

BORKEN-KLEEFELD J, CHEN Y. New emission deterioration rates for gasoline cars - Results from long-term measurements. Atmospheric Environment. 2015; 101:58–64.

BRIGGS DJ, et al. Effects of travel mode on exposures to particulate air pollution. Environment International. 2008; 34:12–22. [PubMed: 17688949]

BROWN DM, et al. Size-dependent proinflammatory effects of ultrafine polystyrene particles: a role for surface area and oxidative stress in the enhanced activity of ultrafines. Toxicology and applied pharmacology. 2001; 175(3):191–199. [PubMed: 11559017]

BRUGGE D, DURANT JL, RIOUX C. Near-highway pollutants in motor vehicle exhaust: A review of epidemiologic evidence of cardiac and pulmonary health risks. Environmental Health. 2007; 6(23)

CHARRON A, HARRISON RM. Primary particle formation from vehicle emissions during exhaust dilution in the roadside atmosphere. Atmospheric Environment. 2003; 37:4109–4119.

CHENG Y-H, LIU Z-S, CHEN C-C. On-road measurements of ultrafine particle concentration profiles and their size distributions inside the longest highway tunnel in Southeast Asia. Atmospheric Environment. 2010; 44:763–772.

CYRYS J, et al. Relationship between indoor and outdoor levels of fine particle mass, particle number concentrations and black smoke under different ventilation conditions. J Expo Anal Environ Epidemiol. 2004; 14(4):275–283. Disponível em: < http://dx.doi.org/10.1038/sj.jea.7500317 >. [PubMed: 15254474]

DAHL A, et al. Traffic-generated emissions of ultrafine particles from pavement–tire interface. Atmospheric Environment. 2006; 40:1314–1323.

DE NAZELLEA, et al. A travel mode comparison of commuters' exposures to air pollutants in Barcelona. Atmospheric Environment. 2012; 59:151–159.

DENNEKAMP M, et al. Exposure to Ultrafine Particles and PM2.5 in Different Micro-environments. Annual of Occupational Hygiene. 2002; 46(1):412–414.

DING H, et al. Analysis of PM2.5 distribution and transfer characteristics in a car cabin. Energy and Buildings. 2016; 127:252–258.

FOX, J., WEISBERG, S. An {R} Companion to Applied Regression. 2. Thousand Oaks, CA: Sage; 2011. Disponível em: < http://socserv.socsci.mcmaster.ca/jfox/Books/Companion >

GOOD N, et al. The Fort Collins Commuter Study: Impact of route type and transport mode on personal exposure to multiple air pollutants. Journal of Exposure Science and Environmental Epidemiology. 2016; 26:397–404. [PubMed: 26507004]

GREENBAUM, DS. SOURCES OF AIR POLLUTION. World Health Organization; 2013. GASOLINE AND DIESEL ENGINES. STRAIF, K.,COHEN, A., *et al*: IARC Scientific Publication No.161

GRIGORATOS, T., MARTINI, G. Brake and tyre wear PM: Literature Review. European Commission; 2014. Non-exhaust traffic related emissions. (EUR 26648 EN)

GROSS, J., LIGGES, U. Tests for Normality. 2015.

HEINRICH, J., et al. Health effects of transport-related air pollution. World Health Organization (WHO); 2005. Studies on health effects of transport-related air pollution; p. 125-183.KRZYZANOWSKI, M.,KUNA-DIBBERT, B., *et al*

HITCHINS J, et al. Concentrations of submicrometre particles from vehicle emissions near a major road. Atmospheric Environment. 2000; 34:51–59.

HUDDA N, et al. Linking in-vehicle ultrafine particle exposures to on-road concentrations. Atmospheric Environment. 2012; 59:578e586. [PubMed: 23888122]

HUDDA N, FRUIN S. Models for predicting the ratio of particulate pollutant concentrations inside vehicles to roadways. Environmental science & technology. 2013; 47(19):11048–11055. [PubMed: 23957386]

HUDDA N, et al. Vehicle and Driving Characteristics That Influence In-Cabin Particle Number Concentrations. Environmental Science & Technology. 2011 Oct 15; 452011(20):8691–8697. Disponível em: < http://dx.doi.org/10.1021/es202025m >. [PubMed: 21928803]

HUEGLIN C, et al. Chemical characterisation of PM2.5, PM10 and coarse particles at urban, near-city and rural sites in Switzerland. Atmospheric Environment. 2005; 39:637–651.

HUSSEIN T, et al. Meteorological dependence of size-fractioned number concentrations of urban aerosol particles. Atmospheric Environment. 2006; 40:1427–1440.

IQSS. [Acesso em: 05-25-2016] Institute for Quantitative Social Science, Harvard University: Multilevel Modeling. 2016. Disponível em: < http://tutorials.iq.harvard.edu/R/Rstatistics/Rstatistics.html#orgheadline36 >

ITO K, THURSTON GD, SILVERMAN RA. Characterization of PM2.5, gaseous pollutants, and meteorological interactions in the context of time-series health effects models. Journal of Exposure Science and Environmental Epidemiology. 2007; 17:S45–S60. [PubMed: 18079764]

JANHÄLL S, et al. Size resolved traffic emission factors of submicrometer particles. Atmospheric Environment. 2004; 38:4331–4340.

KAMINSKY JA, et al. In-Cabin Commuter Exposure to Ultrafine Particles on Commuter Roads in and around Hong Kong’s Tseung Kwan O Tunnel. Aerosol and Air Quality Research. 2009; 9:353–357.

KARAKOÇAK BB, et al. Biocompatibility of gold nanoparticles in retinal pigment epithelial cell line. Toxicology in Vitro. 2016; 37:61–69. [PubMed: 27599945]

KAUR S, NIEUWENHUIJSENB MJ, COLVILE RN. Fine particulate matter and carbon monoxide exposure concentrations in urban street transport microenvironments: a review. Atmospheric Environment. 2007; 41:4781–4810.

KEUKEN, M., et al. Health effects of transport-related air pollution. World Health Organization (WHO); 2005. Contribution of traffic to levels of ambient air pollution in Europe; p. 53-80.KRZYZANOWSKI, M.,KUNA-DIBBERT, B., *et al*: WHO Library Cataloguing in Publication Data

KINGHAM S, et al. Variations in exposure to traffic pollution while travelling by different modes in a low density, less congested city. Environmental Pollution. 2013; 181:211–218. [PubMed: 23871818]

KITTELSON DB, WATTS WF, JOHNSON JP. Nanoparticle emissions on Minnesota highways. Atmospheric Environment. 2004; 38:9–19.

KNIBBS LD, COLE-HUNTER T, MORAWSKA L. A review of commuter exposure to ultrafine particles and its health effects. Atmospheric Environment. 2011; 45:2611–2622.

KNIBBS LD, DE DEAR RJ, MORAWSKA L. Effect of Cabin Ventilation Rate on Ultrafine Particle Exposure Inside Automobiles. Environ Sci Technol. 2010; 44:3546–3551. [PubMed: 20369882]

KRASENBRINK, A., et al. Health effects of transport-related air pollution. World Health Organization (WHO); 2005. Factors determining emissions in the WHO European Region; p.

7-52.KRZYZANOWSKI, M.,KUNA-DIBBERT, B., *et al*: WHO Library Cataloguing in Publication Data

KUHN T, BISWAS S, SIOUTAS C. Diurnal and seasonal characteristics of particle volatility and chemical composition in the vicinity of a light-duty freeway. Atmospheric Environment. 2005; 39:7154–7166.

KULKARNI P, QI C, FUKUSHIMA N. Development of Portable Aerosol Mobility Spectrometer for Personal and Mobile Aerosol Measurement. Aerosol Science and Technology. 2016; 50:1167–1179.

LAMORENA RB, LEE W. Influence of ozone concentration and temperature on ultra-fine particle and gaseous volatile organic compound formations generated during the ozone-initiated reactions with emitted terpenes from a car air freshener. Journal of Hazardous Materials. 2008; 158:471–477. [PubMed: 18336999]

LEAVEY, A. Ph.D. School of Earth, Atmospheric and Environmental Sciences, University of Manchester; Manchester: 2009. Insights into the variables controlling human exposure to ultrafine particle concentrations in urban affinity zones; p. 397

LEAVEY A, et al. Comparison of Measured Particle Lung-Deposited Surface Area Concentrations by an Aerotrak 9000 Using Size Distribution Measurements for a Range of Combustion Aerosols. Aerosol Science and Technology. 2013; 47:966–978.

LIM SS, et al. A comparative risk assessment of burden of disease and injury attributable to 67 risk factors and risk factor clusters in 21 regions, 1990–2010: a systematic analysis for the Global Burden of Disease Study 2010. The Lancet. 2012; 380:2224–60.

LONGLEY ID, et al. Using NOx and CO monitoring data to indicate fine aerosol number concentrations and emission factors in three UK conurbations. Atmospheric Environment. 2005; 39:5157–5169.

LOOMIS D, GROSSE Y, LAUBY-SECRETAN BEA. The carcinogenicity of outdoor air pollution. The Lancet Oncology. 2013; 14(13):1262–1263. [PubMed: 25035875]

LÜDECKE, D. [Acesso em: 05-23-2016] Data Transformation and Labelled Data Utility Functions. 2016a. Disponível em: < https://github.com/sjPlot/sjmisc >

LÜDECKE, D. [Acesso em: 05-24-16] sjPlot: Data Visualization for Statistics in Social Science. 2016b. Disponível em: < https://cran.r-project.org/web/packages/sjPlot/index.html >

MCKENZIE, B. American Community Survey Reports. US Department of Commerce. Economics and Statistics Administration. US Census Bureau; 2015. Who Drives to Work? Commuting by Automobile in the United States: 2013; p. 1-28.(ACS-32)

NCDC, N. C. F. E. I. Quality Controlled Local Climatological Data (QCLCD). 2015.

OBERDORSTER G. Significance of particle parameters in the evaluation of exposure-dose-response relationships of inhaled particles. Inhalation toxicology. 1996; 8:73–89. [PubMed: 11542496]

OSBORNE J, WATERS E. Four assumptions of multiple regression that researchers should always test. Practical Assessment, Research & Evaluation. 2002; 8(2) Disponível em: < http://PAREonline.net/getvn.asp?v=8&n=2 >.

PALMGREN F, et al. Characterisation of particle emissions from the driving car fleet and the contribution to ambient and indoor particle concentrations. Physics and Chemistry of the Earth. 2003; 28:327–334.

PIRJOLA L, et al. Dispersion of particles and trace gases nearby a city highway: Mobile laboratory measurements in Finland. Atmospheric Environment. 2006; 40:867–879.

QI C, KULKARNI P. Miniature Differential Mobility Analyzer for Compact Field-Portable Spectrometers. Aerosol Science and Technology. 2016; 50:1145–1154.

QUEROL X, et al. PM10 and PM2.5 source apportionment in the Barcelona Metropolitan area, Catalonia, Spain. Atmospheric Environment. 2001; 35:6407–6419.

RAGETTLI MS, et al. Commuter exposure to ultrafine particles in different urban locations, transportation modes and routes. Atmospheric Environment. 2013; 77:376–384.

RIEDIKER M, et al. Particulate Matter Exposure in Cars is Associated with Cardiovascular Effects in Healthy Young Men. Am J Respir Crit Care Med. 2004; 169:934–940. [PubMed: 14962820]

RUSSELL, A. Air Pollution and Cancer. World Health Organization; 2013. COMBUSTION EMISSIONS. STRAIF, K.,COHEN, A., *et al*: IARC Scientific Publication No.161

SARNAT JA, et al. Exposure to traffic pollution, acute inflammation and autonomic response in a panel of car commuters. Environmental Research. 2014; 133:66–76. [PubMed: 24906070]

SHI JP, KHAN AA, HARRISON RM. Measurements of ultrafine particle concentration and size distribution in the urban atmosphere. The Science of the Total Environment. 1999; 235:51–64.

SIEGMANN P, et al. A probabilistic source attribution model for nanoparticles in air suspension applied on the main roads of Madrid and Mexico City. Atmospheric Environment. 2008; 42:3937–3948.

SUAREZ L, et al. Personal exposure to particulate matter in commuters using different transport modes (bus, bicycle, car and subway) in an assigned route in downtown Santiago, Chile. Environ Sci: Processes Impacts. 2014; 16:1309–1317.

TAI APK, MICKLEY LJ, JACOB DJ. Correlations between fine particulate matter (PM2.5) and meteorological variables in the United States: Implications for the sensitivity of PM2.5 to climate change. Atmospheric Environment. 2010; 44:3976–3984.

THORPE A, HARRISON RM. Sources and properties of non-exhaust particulate matter from road traffic: A review. Science of the Total Environment. 2008; 400:270–282. [PubMed: 18635248]

USDOT, U. D. O. T. [Acesso em: FHWA-PL-ll-022] SUMMARY OF TRAVEL TRENDS: 2009 National Household Travel Survey; 2009. p. 1-82.Disponível em: < file:///C:/Users/aleavey/Documents/Commuting_Study/Literature/US%20DOT.%20Summary%20of%20travel%20trends%20in%20the%20US.pdf >

USEPA, U. E. P. A. Air Quality System Data Mart internet database. 2015.

VAN WIJNENJH, et al. The exposure of cyclists, car drivers and pedestrians to traffic-related air pollutants. International Archives of Occupational and Environmental Health. 1995; 67(3):187–193. EISSN 1432-1246. [PubMed: 7591177]

WEHNER B, et al. Particle number size distributions in a street canyon and their transformation into the urban-air background: measurements and a simple model study. Atmospheric Environment. 2002; 36:2215–2223.

WEICHENTHAL S, et al. In-Vehicle Exposures to Particulate Air Pollution in Canadian Metropolitan Areas: The Urban Transportation Exposure Study. Environ Sci Technol. 2015; 49:597–605. [PubMed: 25469563]

WEIJERS EP, et al. Variability of particulate matter concentrations along roads and motorways determined by a moving measurement unit. Atmospheric Environment. 2004; 38:2993–3002.

WESTERDAHL D, et al. Mobile platform measurements of Ultrafine particles and associated pollutant concentrations on freeways and residential streets in Los Angeles. Atmospheric Environment. 2005; 39:3597–3610.

WICKHAM, H. ggplot2: Elegant Graphics for Data Analysis. Springer-Verlag; New York: 2009. Disponível em: < http://ggplot2.org >

WINTER, B. Linear models and linear mixed effects models in R with linguistic applications. 2013. Disponível em: < http://arxiv.org/pdf/1308.5499.pdf >

XU B, LIU S, ZHU Y. Ultrafine particle penetration through idealized vehicle cracks. Journal of Aerosol Science. 2010; 41(9):859–868. Disponível em: < http://www.sciencedirect.com/science/article/pii/S0021850210001205 >.

YAMADA H, HAYASHI R, TONOKURA K. Simultaneous measurements of on-road/in-vehicle nanoparticles and NOx while driving: Actual situations, passenger exposure and secondary formations. Science of the Total Environment. 2016; 563–564:944–955.

YAO X, et al. Size distributions and condensation growth of submicron particles in on-road vehicle plumes in Hong Kong. Atmospheric Environment. 2007; 41(16):3328–3338.

ZHANG KM, WEXLER AS. Evolution of particle number distribution near roadways – Part 1: analysis of aerosol dynamics and its implication for engine emission measurement. Atmospheric Environment. 2004; 38:6643–6653.

ZHU Y, et al. In-Cabin Commuter Exposure to Ultrafine Particles on Los Angeles Freeways. ENVIRONMENTAL SCIENCE & TECHNOLOGY. 2007 Apr 01; 412007(7):2138–2145. Disponível em: < http://dx.doi.org/10.1021/es0618797 >. [PubMed: 17438754]

ZHU Y, HINDS WC. Predicting particle number concentrations near a highway based on vertical concentration profile. Atmospheric Environment. 2005; 39:1557–1566.

ZHU Y, et al. Penetration of freeway ultrafine particles into indoor environments. Aerosol Science. 2005; 36:303–322.

ZUURBIER M, et al. Commuters' Exposure to Particulate Matter Air Pollution Is Affected by Mode of Transport, Fuel Type, and Route. Environmental Health Perspectives. 2010; 118(6):783–89. [PubMed: 20185385]

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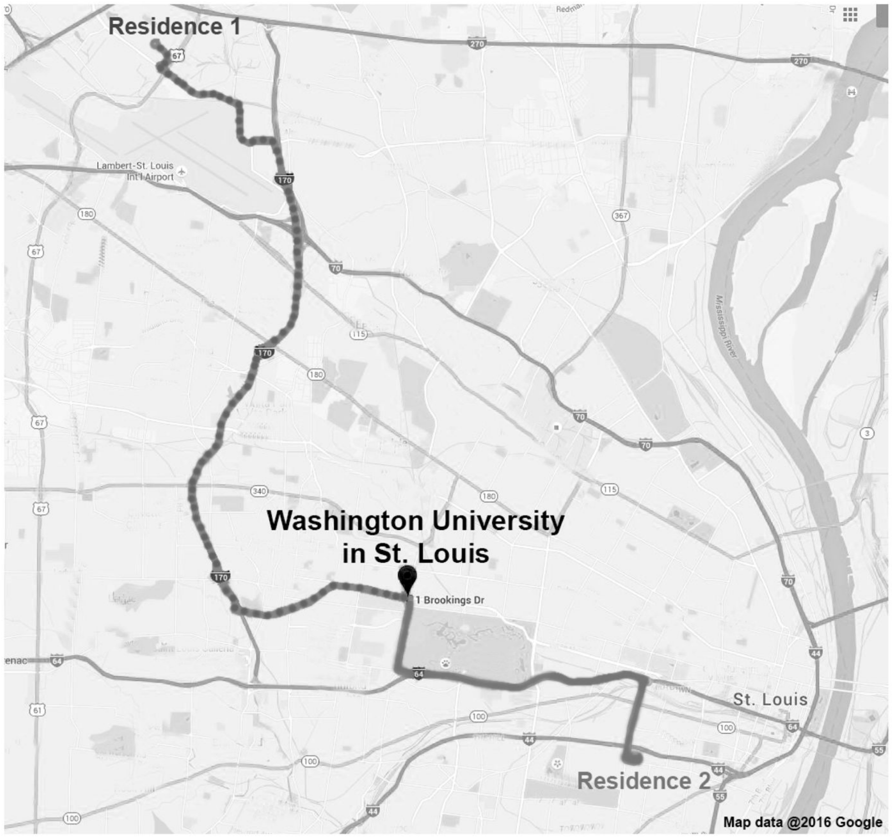


Figure 1.
Map of route travelled during measurement campaign

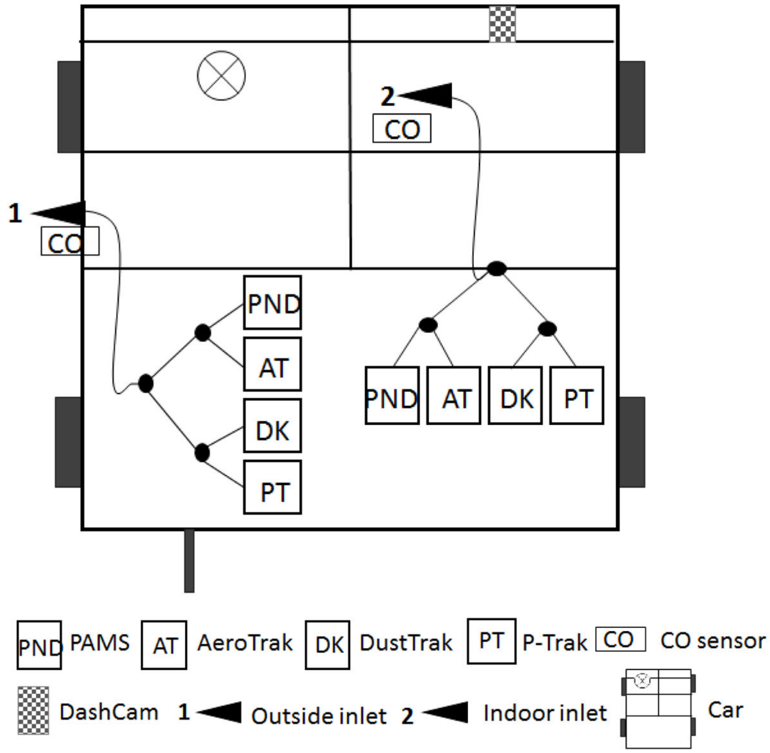
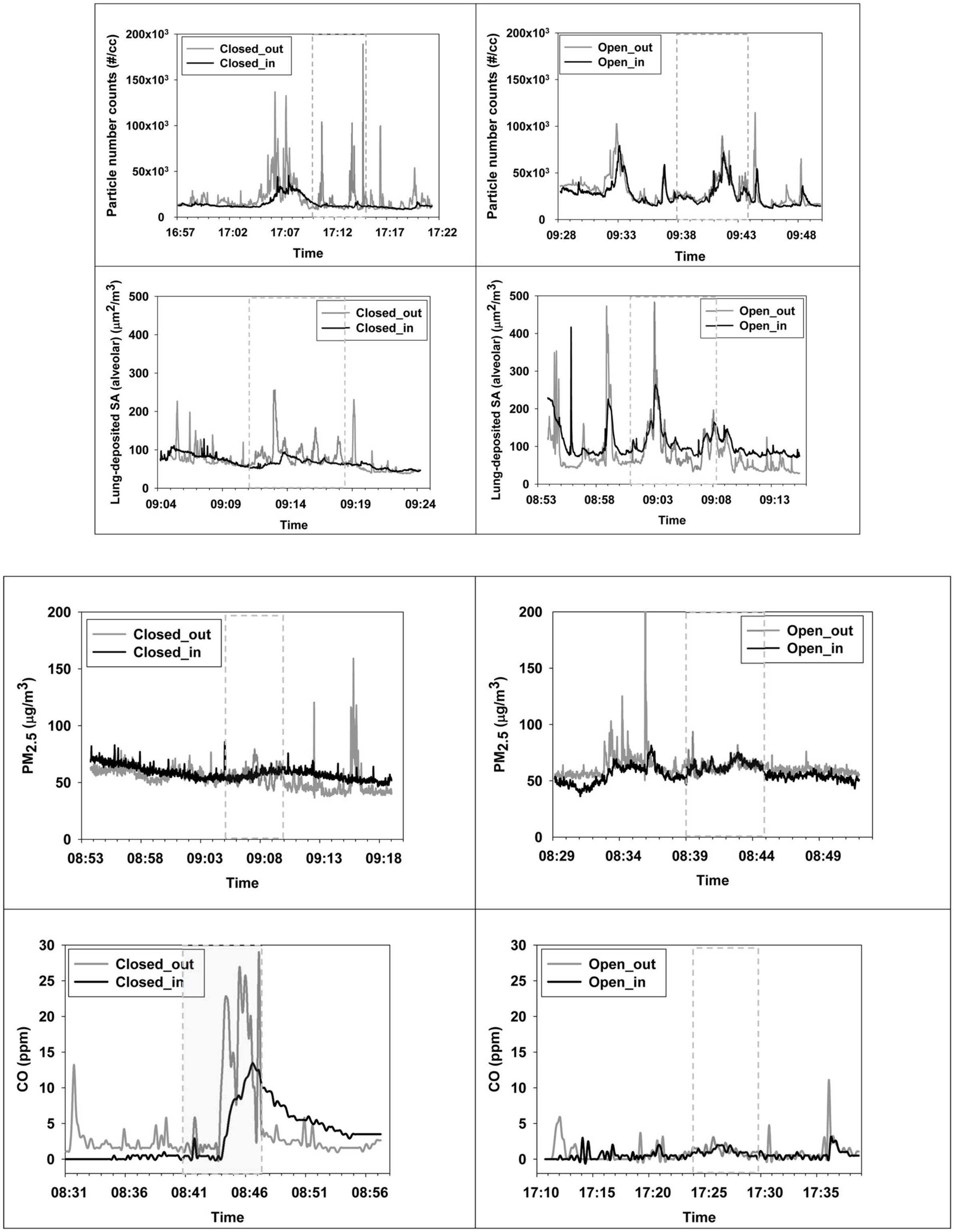


Figure 2.

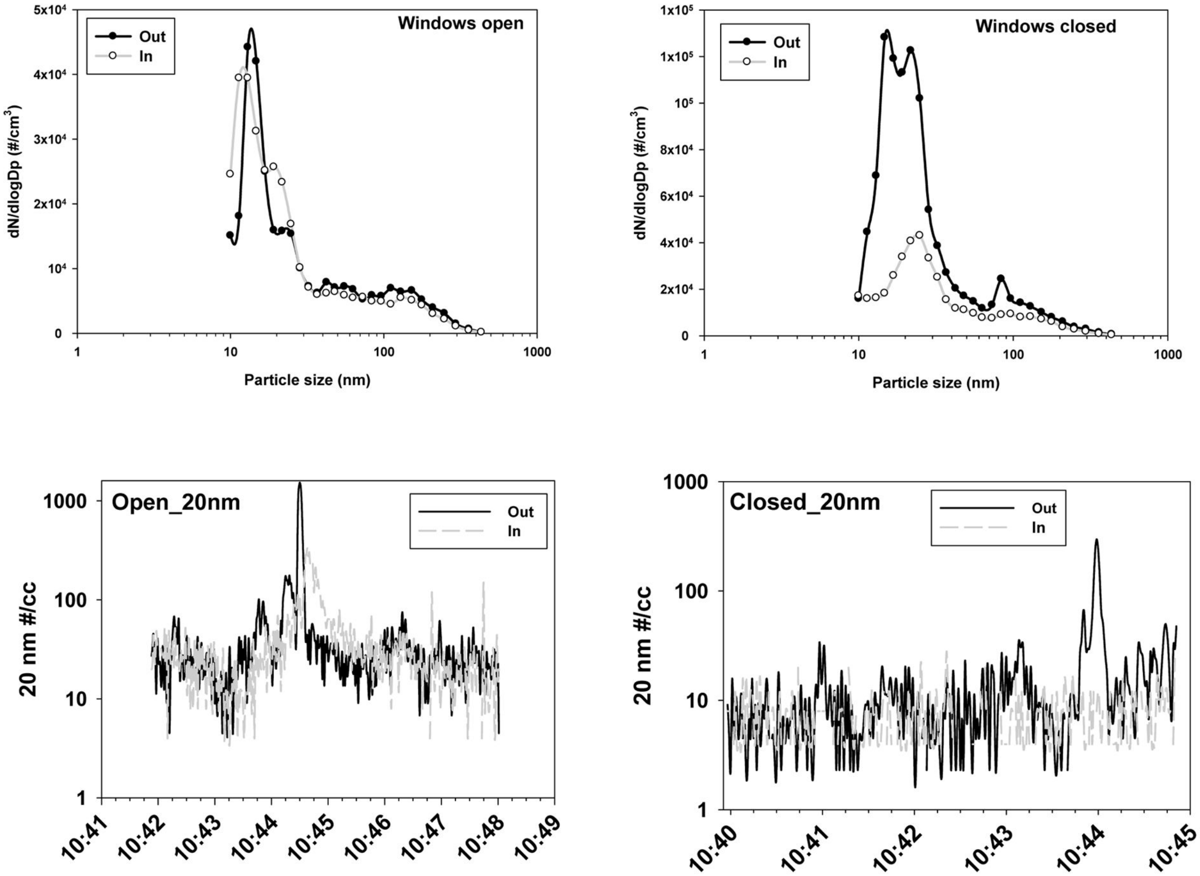
Schematic of the experimental setup.



Particle number counts (#/cc)
200x10³
150x10³
100x10³
50x10³
0
16:57 17:02 17:07 17:12 17:17 17:22
Time
Closed_out
Closed_in
Particle number counts (#/cc)
200x10³
150x10³
100x10³
50x10³
0
09:28 09:33 09:38 09:43 09:48
Time
Open_out
Open_in
Lung-deposited SA (alveolar) (μm²/m³)
500 400 300 200 100 0
09:04 09:09 09:14 09:19 09:24
Time
Closed_out
Closed_in
Lung-deposited SA (alveolar) (μm²/m³)
500 400 300 200 100 0
08:53 08:58 09:03 09:08 09:13
Time
Open_out
Open_in
PM2.5 (μg/m³)
200 150 100 50 0
08:53 08:58 09:03 09:08 09:13 09:18
Time
Closed_out
Closed_in
PM2.5 (μg/m³)
200 150 100 50 0
08:29 08:34 08:39 08:44 08:49
Time
Open_out
Open_in
CO (ppm)
30 25 20 15 10 5 0
08:31 08:36 08:41 08:46 08:51 08:56
Time
Closed_out
Closed_in
CO (ppm)
30 25 20 15 10 5 0
17:10 17:15 17:20 17:25 17:30 17:35
Time
Open_out
Open_in

Figure 3(a–h).

Time-series depicting simultaneous in-car / outdoor concentrations during windows open and windows closed for (a–b) particle number counts, (c–d) lung-deposited SA, (e–f) $PM_{2.5}$, and (g–h) CO. The shaded rectangle is the time spent on the freeway


Windows open
Out
In
dN/dlogDp (#/cm3)
Particle size (nm)
Windows closed
Out
In
dN/dlogDp (#/cm3)
Particle size (nm)
Open_20nm
Out
In
20 nm #/cc
Closed_20nm
Out
In
20 nm #/cc

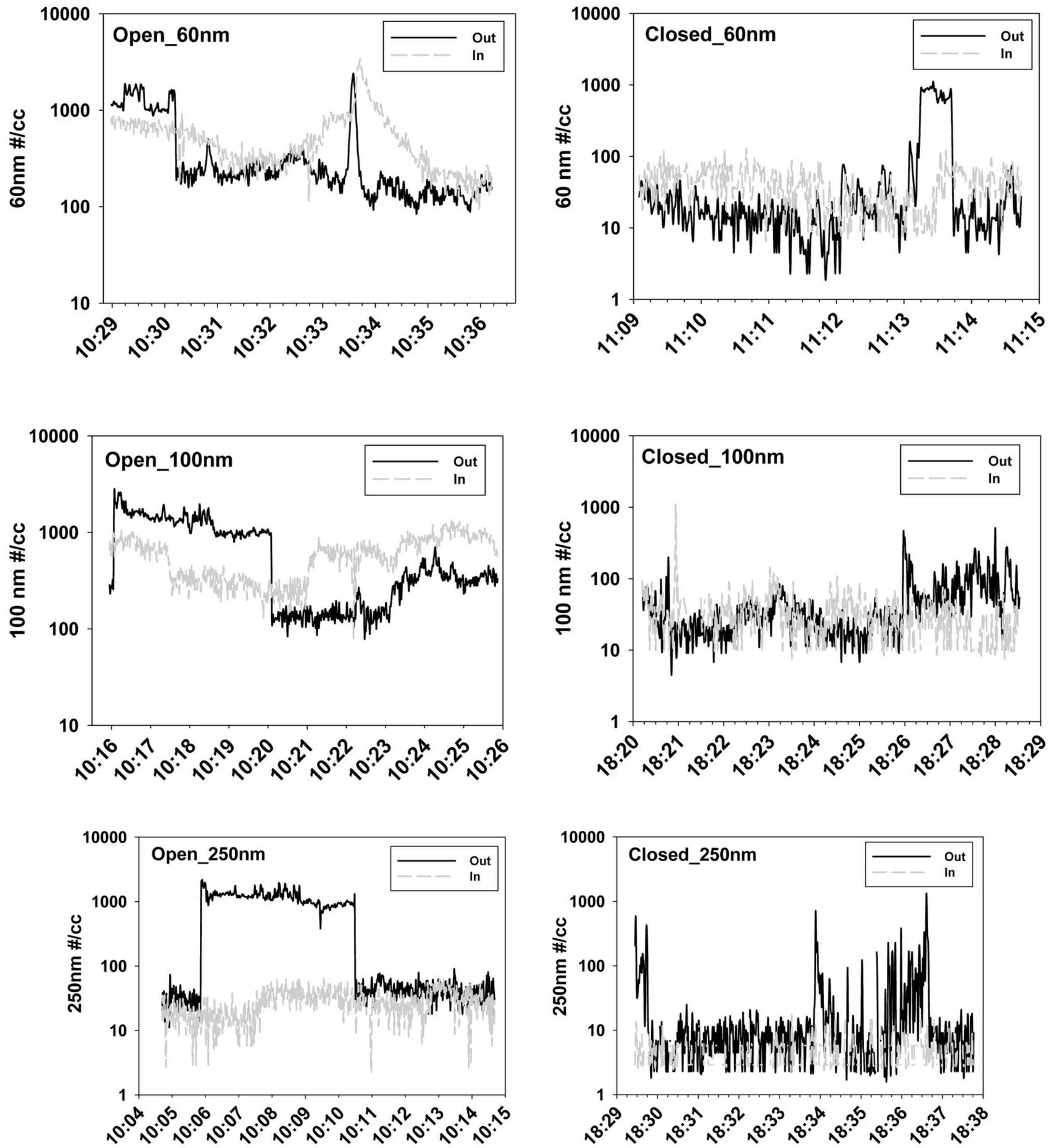
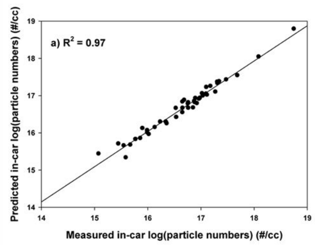
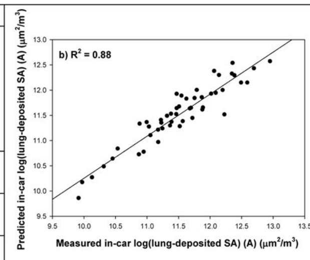


Figure 4(a–j).

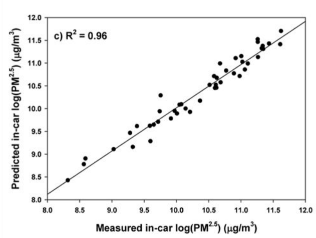
Simultaneous in-car / outdoor concentrations during windows open and closed for (a–b) PNSD, (c–d) 20 nm, (e–f) 60 nm, and (g–h) 100 nm, (i–j) 250 nm.



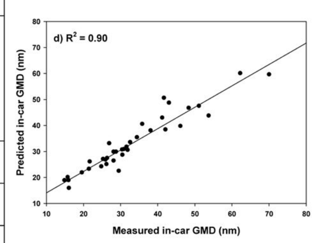
Fixed-effects	b	SE
Intercept[1]	-1.512	1.38
lnPT_out**	1.089	0.08
AC**	-0.416	0.10
Closed**	-0.563	0.08
Fan*	-0.217	0.10
Random-effects	**Var**	**SD**
Day	0.135	0.37
Residual	0.026	0.16
Random intercepts -0.65 to +1.15 about the mean		



Fixed-effects	b	SE
Intercept[1]	3.715	1.34
lnAR_out**	0.461	0.12
AC*	-0.331	0.14
Closed**	-0.328	0.11
Fan	-0.124	0.13
Temp**	0.040	0.01
Random-effects	**Var**	**SD**
Day	0.06	0.25
Residual	0.08	0.28
Random intercepts -0.39 to +0.31 about the mean		



Fixed-effects	b	SE
Intercept[1]	0.330	0.45
lnDK_out**	0.983	0.04
AC*	-0.218	0.09
Closed	-0.087	0.08
Fan	-0.081	0.09
Random-effects	**Var**	**SD**
Day	0.007	0.08
Residual	0.036	0.19
Random intercepts -0.09 to +0.09 about the mean		



Fixed-effects	b	SE
Intercept[1]	1.141	6.19
GMD_out**	0.841	0.17
AC	1.333	2.05
Closed**	4.314	1.83
Fan	-2.160	1.74
Random-effects	**Var**	**SD**
Day	85.73	9.26
Residual	28.49	5.34
Random intercepts -11.87 to 21.84 about the mean		

Figure 5(a–d).

Sensitivity models comparing measured and predicted in-car data from LME analysis for: a) particle number concentration, b) lung-deposited SA (A), c) $PM_{2.5}$, and d) GMD.

* = statistically significant at the 95th CI; ** = statistically significant at the 99th CI; all dependent variables, with the exception of GMD, were log-transformed, hence all coefficients are for the log of the data; *b* = estimates of regression coefficients; SE = standard error; Var = variance; SD = standard deviation. Windows open is the reference level for the categorical ventilation variable. [1]The intercept for the mean of the explanatory variables which is also the mean of all random intercepts.

Table 1

Variables for whole journey outdoor concentrations (log-transformed area under the curve)

Variable	Pollutant metric														
	Particle numbers (#/cc)			PM2.5 ($\mu g/m^3$)			Lung-deposited SA ($\mu m^2/m^3$)			CO (ppm)			GMD (nm)		
	b	**p**	**adj-R^2**	**b**	**p**	**adj-R^2**	**b**	**p**	**adj-R^2**	**b**	**p**	**adj-R^2**	**b**	**p**	**adj-R^2**
log WindSp	−0.333	0.05	0.25	-	-	-	−0.362	0.02	0.28	-	-	-	−4.868	0.07	0.05
Temp	−0.014	0.05	0.18	0.032	<0.01	0.13	-	-	-	-	-	-	0.306	0.02	0.18
RH	0.017	<0.01	0.25	0.029	<0.01	0.09	0.016	<0.01	0.18	-	-		-	-	-
DewPoint	-	-	-	0.053	<0.01	0.44	0.026	<0.01	0.26	0.090	0.01	0.21	-	-	
WindDir															
W*	16.25	<0.01	0.17	9.634	<0.01	0.29	10.03	<0.01	0.21	-	-	-	-	-	-
E	0.253	0.19		0.850	<0.01		0.493	<0.01		-	-		-	-	
N	0.140	0.50		0.440	0.12		0.275	0.15		-	-		-	-	
S	0.354	<0.01		0.618	0.02		0.376	0.02		-	-		-	-	
log NO_x ^	0.334	<0.01	0.36	0.437	<0.01	0.15	0.368	<0.01	0.24	-	-	-	-	-	-
log $PM_{2.5}$ ^	0.354	0.01	0.18	0.622	<0.01	0.33	0.389	<0.01	0.27	1.191	0.02	0.16	-	-	
log CO ^	0.599	<0.01	0.27	0.867	<0.01	0.13	0.731	<0.01	0.31	-	-	-	-	-	
AM*	16.63	<0.01	0.25	9.914	<0.01	0.07	10.52	<0.01	0.12	2.586	0.12	0.08	30.57	<0.01	0.04
PM	−0.403	<0.01		−0.611	<0.01		−0.371	<0.01		−1.447	<0.01		2.587	0.08	
TimeBB	-	-	-	0.003	<0.01	0.12	0.002	0.01	0.10	0.008	0.04	0.13	-	-	-

This table presents all continuous and categorical explanatory variables that were significant at the 90^{th}, 95^{th}, or 99^{th} % CI. The metric used to present all dependent variables (except GMD) was area under the curve. These data were log-transformed to meet normality assumptions. Because of this metric, all regressions were controlled for the duration of a journey, so that any bias is removed.

GMD = geometric mean diameter. # = number;

* = the reference level of the categorical variable;

b = estimates of regression coefficient; p = p-value of b; adj-R^2 = coefficient of determination.

^Measured at a fixed-site monitor, part of the EPA distribution network, and log-transformed to meet normality assumptions.